GENERALIZATIONS OF THE LANDAU-ZENER THEORY IN THE PHYSICS OF NANOSCALE SYSTEMS

A Dissertation

by

NIKOLAI SINITSYN

Submitted to the Office of Graduate Studies of
Texas A&M University
in partial fulfillment of the requirements for the degree of

DOCTOR OF PHILOSOPHY

May 2004

Major Subject: Physics

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Approved as to style and content by:

Valery L. Pokrovsky (Chair of Committee)	Roland E. Allen (Member)
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ABSTRACT

Generalizations of the Landau-Zener Theory in the Physics of Nanoscale Systems.

(May 2004)

Nikolai Sinitsyn, B.S., Belarus State University

Chair of Advisory Committee: Dr. Valery L. Pokrovsky

Nanoscale systems have sizes intermediate between atomic and macroscopic ones. Therefore their treatment often requires a combination of methods from atomic and condensed matter physics. The conventional Landau-Zener theory, being a powerful tool in atomic physics, often fails to predict correctly nonadiabatic transition probabilities in various nanostructures because it does not include many-body effects typical for mesoscopics. In this research project the generalizations of the Landau-Zener theory that solve this problem were studied. The multistate, multiparticle and nonunitary extensions of the theory have been proposed and investigated. New classes of exactly solvable models have been derived. I discuss their applications in problems of the molecular condensate dissociation and of the driven charge transport. In application to the physics of nanomagnets new approaches in modeling the influence of the environment on the Landau-Zener evolution are proposed and simple universal formulas are derived for the extensions of the theory that include the coupling to noise and the nuclear spin bath.

To my wife Marina and my mama Luba

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My progress at Texas A&M would not have been possible without my previous teachers at Belarus State University: Professors Eugeny Ushakov, Artur Tregubovich, Andrey Lavrinenko, Vyacheslav Kuvshinov, Leonid Markovich and my school physics and mathematics teachers: Sergei Mikhailovich Karpuk and Natalia Vladimirovna Gurinovich. The support from my brother Vladimir is invaluable.

My mother and my beloved wife Marina always inspire and motivate me. I am happy to dedicate this dissertation to them.

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CHAPTER I

INTRODUCTION

The Landau-Zener formula [1, 2] for transition probabilities at avoided crossing of two quantum levels is one of a few most fundamental results of non-stationary quantum mechanics. Its rather general character and simplicity makes it extremely suitable for versatile applications.

Traditionally it was employed in quantum chemistry [3] and in collision theory [4, 5]. A recent treatment of the experiments on the quantum molecular hysteresis in nanomagnets by Wernsdorfer and Sessoli [6, 7, 8] was a real triumph of the LZ theory. Landau-Zener formula and its generalizations have been recently employed also in various problems related to the charge transport in nanostructures [9, 10, 11, 12, 13, 14], Bose-Einstein condensates [15, 16, 17, 18, 19, 20, 21, 22, 23] and quantum computing [24, 25].

Emerging applications in mesoscopic and nanoscale systems stimulated recent theoretical progress in the Landau-Zener (LZ) theory. Mesoscopic systems have sizes intermediate between atomic and macroscopic ones. Therefore their treatment often requires a combination of methods from atomic and condensed matter physics. The conventional Landau-Zener theory, being a powerful tool in atomic physics, often fails to predict correctly nonadiabatic transition probabilities in various nanostructures because it does not include many-body effects typical for mesoscopics. In order to include various many-body interactions, the new branches of the theory such as nonlinear, nonunitary, multistate and multiparticle Landau-Zener models have been introduced [19, 20, 15, 21, 26, 27, 28].

The journal model is *Physical Review Letters*.

Generally it is very difficult to obtain analytic results for Schrödinger equations with time-dependent Hamiltonians. The most general form of the time-dependent Shrödinger equations for two states is

$$\begin{aligned} i\dot{b}_1 &= E_1(t)b_1 + \Delta(t)b_2 \\ i\dot{b}_2 &= E_2(t)b_2 + \Delta(t)b_1 \end{aligned} \tag{1.1}$$

Functions $E_1(t)$ and $E_2(t)$ are called diabatic energies. There is no known general solution of the system (1.1); however, according to the adiabatic theorem, transitions between two states are strongly suppressed for a sufficiently large energy difference between states. The adiabaticity is violated when a pair of levels move toward each other strongly enhancing the transitions between the two states near the crossing point of energy curves. Landau and Zener determined the transition probabilities in the special case of the two-level crossing, which can be employed as an approximation in more general situations.

Near a crossing point the dependence of energy levels on time is approximately linear $E_\alpha(t) = \dot{E}_\alpha t;\ \alpha = 1, 2$, whereas the non-diagonal matrix elements of the Hamiltonian can be taken constants. In terms of new amplitudes $a_{1,2} = e^{-i(\dot{E}_1+\dot{E}_2)t^2/4}b_{1,2}$, after a time rescaling, equations (1.1) can be simplified as follows:

$$\begin{aligned} i\dot{a}_1 &= ta_1 + \gamma a_2 \\ i\dot{a}_2 &= -ta_2 + \gamma a_1 \end{aligned} \tag{1.2}$$

where $\gamma = \sqrt{2\Delta/\dot{\Omega}},\ \dot{\Omega} = \dot{E}_1 - \dot{E}_2$. This type of two level crossing was studied and solved already in 1932 by Landau [1], Zener [2], Stückelberg [29] and Majorana [30].

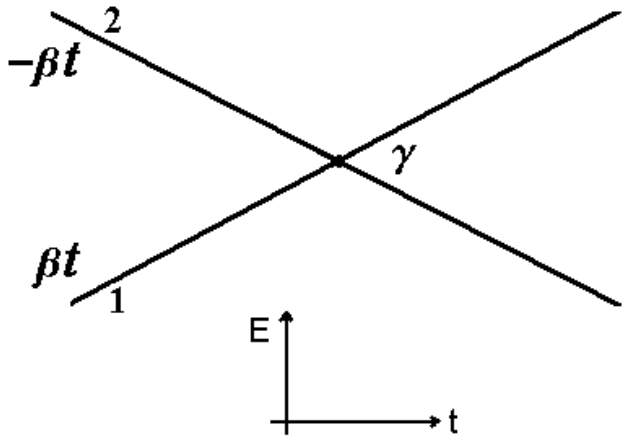

Fig. 1. Diabatic energies in the two-state Landau-Zener model. γ is the strength of the coupling between levels and β shows how fast levels cross each other.

It is convenient to visualize parameters of the corresponding Hamiltonian in a time-energy picture like Fig.1. Eliminating a_2 from these equations, we find the parabolic cylinder equation for $a_1(t)$. Its solution which has asymptotics $a_1 \simeq \exp(-\frac{it^2}{2} - i\gamma^2 \ln|t|)$ and $a_2 = 0$ at $t \to -\infty$ is the Weber function $D_{-i\gamma^2}(\sqrt{2}e^{i\pi/4}t)$ whose asymptotics at $t \to \infty$ are well known [31]. The scattering matrix for the two level system is conveniently written in terms of modified amplitudes $c_1 = a_1 \exp(if)$; $c_2 = a_2 \exp(-if)$ where $f = \frac{t^2}{2} + \gamma^2 \ln|t|$. It reads:

$$U_\infty = \begin{pmatrix} \exp(-\frac{\pi\gamma^2}{2}) & -\frac{\sqrt{\pi}\exp(-\frac{\pi\gamma^2}{4}+\frac{i\pi}{4})}{\gamma\Gamma(-\frac{i\gamma^2}{2})} \\ \frac{\sqrt{\pi}\exp(\frac{\pi\gamma^2}{4}-\frac{i\pi}{4})}{\gamma\Gamma(\frac{i\gamma^2}{2})} & \exp(-\frac{\pi\gamma^2}{2}) \end{pmatrix} \tag{1.3}$$

Having the expression for the amplitudes it is straightforward to find the transition probability from one state into another

$$P_{LZ} = 1 - \exp\left(-\pi|\gamma|^2\right) \tag{1.4}$$

Extensions of the LZ theory to the case of multilevel crossing are less general.

Nevertheless, some of them were realistic enough to justify remarkable efforts on the side of theorists for their analysis. The pioneering work by Demkov and Osherov [32] treated exactly the crossing of a single level with a band of parallel levels. In the work [33] Hioe and Carrol solved a problem of transitions in a Zeeman multiplet of an arbitrary spin S in a magnetic field with a constant perpendicular component and a time-dependent parallel component passing through zero value. Level correlations and localization in energy space were studied in [34]. Numerous generalizations of these results were found [28, 35, 36, 37, 38, 39, 40, 41, 42, 43].

Despite a strong progress during the last decade the multistate Landau-Zener problem remains not well understood. One of the striking facts about it is the existence of very general empirical formulas for some elements of the transition probability matrix. Such formulas still remain unproved although they have been confirmed by multiple numerical tests. Many other recent results support the idea that strong progress in an exact treatment of those models is possible.

To apply the LZ formula and its multi-state extensions to real systems it is often necessary to take into account the interaction with environment. Such attempts were made in a series of works [44, 45, 46, 47, 48, 49, 50, 51, 52, 53, 54, 55, 56, 57], however the problem was not solved completely. Kayanuma *et al.* [44, 45, 46] have obtained an elegant analytic result for the diagonal white noise. The non-diagonal colored noise was considered by Kayanuma [47] for the two-level crossing without a constant coupling term. He has found the transition probability in the limit of an infinitely short noise correlation time and for very special type of noise correlator. We performed a systematic study of the influence of noise, including colored noise, onto the LZ transitions and generalized it to multistate LZ problems. One of the main results is the derivation of the most general formula for the transition probability in a system coupled to fast stationary noise with arbitrary form of the noise correlator

and taking into account the regular transverse field.

Interaction with environment cannot be always reduced to interaction with a noisy field. This is the case of many molecular nanomagnets whose electronic spin is coupled to the nuclear spin bath. Due to strong interaction with the electronic spin of a nanomagnet, dynamics of the nuclear spins are strongly influenced by the dynamics of the central system and hence the fluctuations of the effective hyperfine field cannot be considered as a stationary noise. Such a problem requires a completely different approach [58, 59, 60]. The most reliable description of such LZ transitions can be done in the framework of the model of incoherent Landau-Zener transitions in the greed of intersecting levels that represent different states of the spin bath [4, 61]. Our numerical simulations demonstrated complete success of this approach [62].

Sometimes dynamics of many-body systems can be described by Landau-Zener-like equations whose physical interpretation can be different from nonadiabatic intersection of quantum energy levels. We study such an example in a Bose condensate which requires generalization of the LZ problem to nonunitary evolution [21, 22, 63].

The plan of this thesis is the following. Chapter II. is devoted to multistate and multiparticle extensions of the Landau-Zener theory. In sections A. and B. of this chapter the contour integral approach to solving the Demokov-Osherov and bow-tie models is reviewed. Section C. reviews semi-empirical formulas, valid for any multistate Landau-Zener model. Sections D. and E. demonstrate another approach to exact treatment of the problem by employing symmetry arguments. In section F. the Landau-Zener theory is applied to the problem of Bose condensate dissociation and the corresponding extension of the theory to the nonunitary evolution is studied. Section G. describes a solvable model with formally an infinite number of states. Chapter III. is devoted to the problem of the Landau-Zener transitions in the presence of an additional noisy field which simulates coupling to a fluctuating environment.

First in section A. I introduce the Bloch tensor technique to treat the evolution of density matrix of an arbitrary spin. Then in sections B. and C. I solve a simplified problem of transitions mediated by a noisy field only (at zero regular transverse field). Sections D. and E. are devoted to the problem of including the regular transverse field into the final result. Sections F. and G. describe limiting cases which require a special treatment. In chapter IV. the application of the Landau-Zener theory to the physics of nanomagnets is reviewed. The role of two distinct mechanisms of spin bath effects are studied analytically in sections A. and B. of this chapter, and section C. shows the result of the direct quantum mechanical simulations which confirmed the main analytical predictions.

CHAPTER II

MULTISTATE LANDAU-ZENER PROBLEM

The simplest and straightforward generalization of the Landau-Zener problem is the problem of finding of the transition amplitudes for a multistate system with the Hamiltonian, whose matrix form reads

$$H = Bt + A, \tag{2.1}$$

where A and B are Hermitian matrices with time independent elements. In the form where matrix B is diagonalized this problem is referred as the multistate Landau-Zener problem. In its general form this problem is still unsolved, but a number of exact results for special choices of the matrices B and A were found [28, 35, 36, 37, 38, 39, 33, 15, 40, 41, 32, 42, 43].

In almost all available exact solutions the transition probabilities are expressed in terms of the genuine two-level LZ formula successively applied at each diabatic level intersection. In other physical problems such a procedure is often applied as an approximation [4, 64, 65]. These problems include atomic and molecular collisions [66] and the transitions at crossing of two Rydberg multiplets of energy levels [67, 68, 69].

It is always possible to find a time independent basis in which the matrix B is diagonal. Denoting the elements of A as $A_{kk} = \epsilon_k$ and $A_{kl} = v_{kl}$ (for $k \neq l$) the Schrödinger equation can be always written in the following form

$$i\frac{d}{dt}\begin{pmatrix} a_1(t) \\ \vdots \\ \vdots \\ a_n(t) \end{pmatrix} = \begin{pmatrix} \epsilon_1 + \beta_1 t & v_{12} & \cdots & v_{1n} \\ v_{21} & \ddots & \ddots & \vdots \\ \vdots & \ddots & \ddots & v_{n-1,n} \\ v_{n1} & \cdots & v_{n,n-1} & \epsilon_n + \beta_n t \end{pmatrix} \begin{pmatrix} a_1(t) \\ \vdots \\ \vdots \\ a_n(t) \end{pmatrix} \tag{2.2}$$

with real constants ϵ_k, β_k and complex constants $v_{lk} = v_{kl}^*$ and $v_{kl} = 0$ whenever $\beta_k = \beta_l$. This is a system of first-order complex linear differential equations. It conserves probability i.e. $\sum_k |a_k(t)|^2 = 1$.

In physical terms one can interpret the equation (2.2) as describing a quantum mechanical system with some number n of states or levels whose unperturbed (diabatic) energies $E_k(t) = \epsilon_k + \beta_k t$ are linear functions of time, and which are coupled by the constant off-diagonal matrix elements v_{kl}; $a_k(t)$ is the amplitude for the system to be in a state k. Except for the trivial case where all eigenvalues of the matrix B are equal, there will be pairs (k, l) of levels, whose diabatic energies $E_k(t)$ and $E_l(t)$ cross at some point in time.

If all the couplings v_{kl} are equal to zero, the time evolution of the system is trivial. The different degrees of freedom are decoupled and the general solution is $a_k(t) = c_k e^{-i(\epsilon_k t + \beta_k t^2/2)}$, with constant initial values c_k. If on the other hand there are non-zero couplings, transitions between different states become possible. However, for large times t the terms $\beta_k t$ on the diagonal in (2.2) will cause more and more rapid relative oscillations between the different amplitudes, and thus suppress the mixing due to the coupling terms, so that amplitudes $a_k(t)$ approach limits for $t \to \pm\infty$, or, in other words, for all k and l the limit

$$|S_{kl}| = \lim_{t' \to +\infty, t \to -\infty} |U_{lk}(t', t)| \tag{2.3}$$

exists (where $U_{lk}(t', t)$ is the time evolution operator for our equation). In physical terms this means that there are well defined transition probabilities $|S_{kl}|^2$ for the system to transfer from a state l at time $t \to -\infty$ to a state k at $t \to +\infty$. Since the transition probabilities are the quantities of physical interest, the task of the multistate Landau-Zener problem is to calculate the S-matrix defined by (2.3).

In spite of the simplicity of the definition, analytic solutions to the equation (2.2)

have so far been found only for some special cases. The following sections contain the discussion of such known solutions.

A. The Demkov-Osherov model

The Demkov-Osherov model (DOM) [32] is the earliest found solvable multistate generalization of the Landau-Zener model. Assume that all but one of the eigenvalues of the matrix B are equal to each other. It means that a single level crosses a band of parallel ones as it is shown in Fig.2. After simple symmetry transformations the corresponding Schrödinger equation can be written in the form

$$i\frac{d}{dt}\begin{pmatrix} a_0(t) \\ a_1(t) \\ \vdots \\ a_n(t) \end{pmatrix} = \begin{pmatrix} \beta t & v_1 & \cdots & v_n \\ v_1^* & \epsilon_1 & \ddots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ v_n^* & 0 & \cdots & \epsilon_n \end{pmatrix} \begin{pmatrix} a_0(t) \\ a_1(t) \\ \vdots \\ a_n(t) \end{pmatrix} \tag{2.4}$$

with $\beta \neq 0$ and ϵ_k ordered according to $\epsilon_1 < \epsilon_2 < \ldots < \epsilon_n$. No one of the ϵ_k are equal and $\beta > 0$.

Equation (2.4) can be solved via the Fourier transformation. After introducing the ansatz $a_k(t) = \int_C d\omega e^{-i\omega t} u_k(\omega)$ (where C is a contour in complex plane) the equations for the functions $u_k(\omega)$ read

$$\begin{gathered} \omega u_0(\omega) = -i\beta \frac{du_0(\omega)}{d\omega} + \sum_{k=1}^{n} v_k u_k(\omega) \\ \omega u_k(\omega) = \epsilon_k u_k(\omega) + v_k^* u_0(\omega), \;\; (k \neq 0) \end{gathered} \tag{2.5}$$

The problem is reduced only to a single differential plus n algebraic equations that

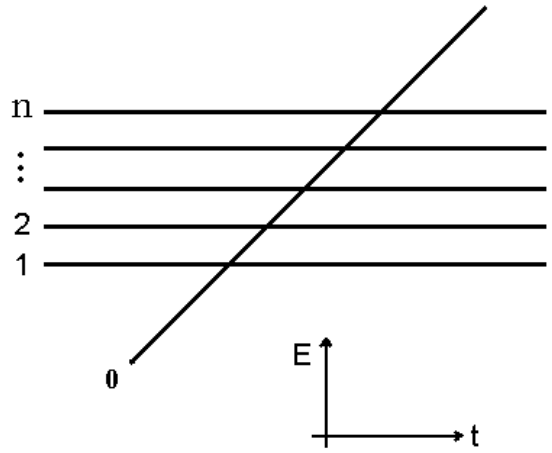

Fig. 2. Time-dependence of diabatic energies in the Demkov-Osherov model. One level crosses a band of other ones having the same slope.

can be trivially solved as

$$u_0(\omega) = \prod_k (\omega - \epsilon_k)^{-iz_k}$$
$$u_k(\omega) = \frac{v_k^*}{\omega - \epsilon_k} u_0(\omega), \quad (k = 1, \ldots, n) \tag{2.6}$$

where $z_k = |v_k|^2/\beta$. Finally, the solution for amplitudes $a_k(t)$ can be found by the inverse Fourier transformation

$$a_k(t) = \int_C d\omega e^{-i(\omega t - \omega^2/(2\beta))} u_k(\omega) \tag{2.7}$$

The contour must be chosen so that the integral (2.7) converges and at times $t \to -\infty$ satisfies the imposed initial conditions. Since only asymptotics are of interest, after finding such a contour, one can suppose that t is a large positive parameter and treat this integral in the stationary phase approximation. Details of those calculations can be found in [35].

Finally, the absolute values of the S-matrix components are [32]:

$$
\begin{aligned}
&|S_{00}| = e^{-\pi(z_1+\ldots+z_n)} \\
&|S_{0l}| = (1-e^{-2\pi z_l})^{1/2} e^{-\pi(z_{l+1}+\ldots+z_n)} \\
&|S_{k0}| = e^{-\pi(z_1+\ldots+z_{k-1})}(1-e^{-2\pi z_k})^{1/2}, (k=1,\ldots,n) \\
&|S_{kl}| = 0, (1 \le k < l) \\
&|S_{ll}| = e^{-\pi z_l} \\
&|S_{kl}| = (1-e^{-2\pi z_l})^{1/2} e^{-\pi(z_{l+1}+\ldots+z_{k-1})}(1-e^{-2\pi z_k})^{1/2}, (k>l)
\end{aligned}
\tag{2.8}
$$

where the index $l = 1 \ldots n$ and $z_k = |\gamma_k|^2/\beta$.

Transition probabilities in DOM do not depend on the ϵ_k for a given ordering. It is quite easy to see that they coincide with predictions of the independent crossing approximation, in which the two-state Landau-Zener formula is applied at a crossing of any pair of diabatic levels in time-ordered sequence.

B. The bow-tie and the generalized bow-tie models

In the bow-tie model all levels are crossing in the same point, and there is one special state, say a_0 coupled to all others, but with those not being coupled to each other. The evolution equation for amplitudes can be written in the following form

$$
i\frac{d}{dt}\begin{pmatrix} a_0(t) \\ a_1(t) \\ \vdots \\ a_n(t) \end{pmatrix} = \begin{pmatrix} 0 & v_1 & \cdots & v_n \\ v_1 & \beta_1 t & \ldots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ v_n & 0 & \cdots & \beta_n t \end{pmatrix} \begin{pmatrix} a_0(t) \\ a_1(t) \\ \vdots \\ a_n(t) \end{pmatrix} \tag{2.9}
$$

As was found in [40], the solution of the bow-tie model can be derived from the solution of the slightly richer model which is called the generalized bow-tie model (GBTM). In the GBTM the number N of states is arbitrary but larger than 2. Two of the diabatic energy levels, labelled by 0^- and 0^+, are horizontal and all others are

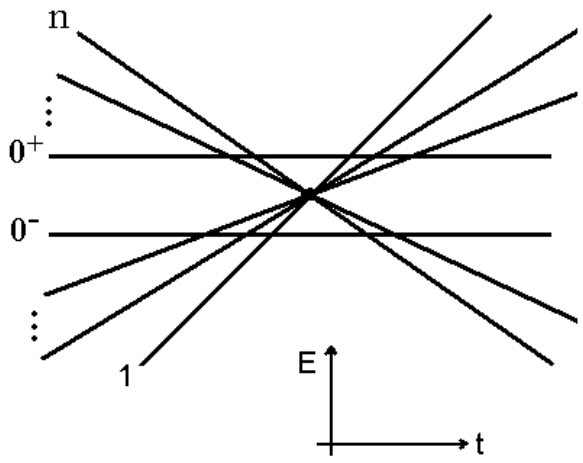

Fig. 3. Time-dependence of diabatic energies in the generalized bow-tie model. All but two levels cross in one point. States 1 to n are not directly coupled to each other, but all of them are coupled to parallel levels 0^- and 0^+.

slanted as it is shown in Fig.3. All $(N-2)$ slanted curves cross at the same instant in time which can be chosen as zero. The energies at which the multiple crossing occurs is naturally chosen as the origin of the energy scale. The energies of the horizontal curves are symmetrical with respect to the origin being shifted by $\pm\epsilon/2$. Hence the evolution equation has the form

$$i\frac{d}{dt}\begin{pmatrix} a_{0^+}(t) \\ a_{0^-}(t) \\ a_1(t) \\ \vdots \\ a_n(t) \end{pmatrix} = \begin{pmatrix} \frac{1}{2}\epsilon & 0 & v_1 & \cdots & v_n \\ 0 & -\frac{1}{2}\epsilon & v_1 & \cdots & v_n \\ v_1 & v_1 & \beta_1 t & \ddots & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ v_n & v_n & 0 & \cdots & \beta_n t \end{pmatrix} \begin{pmatrix} a_{0^+}(t) \\ a_{0^-}(t) \\ a_1(t) \\ \vdots \\ a_n(t) \end{pmatrix} \tag{2.10}$$

Solution of (2.10) can be carried out like the solution of DOM using the contour integral method. Straightforward application of the Fourier transformation leads to a second order differential equation which is not so simple to investigate. Instead, first one can make a change of variables so that after Fourier transformation the resulting

differential equation is of the first order. Lets introduce

$$b_1(t) = \frac{1}{\sqrt{2}}(a_{0^+}(t) + a_{0^-}(t)), \quad b_2(t) = \frac{1}{\sqrt{2}}(a_{0^+}(t) - a_{0^-}(t)) \tag{2.11}$$

In the limit $\epsilon = 0$ the state $b_2(t)$ decouples from all others and resulting evolution equations coincide with normal bow-tie model (2.9), so that we can consider only the GBTM and if necessarily set $\epsilon = 0$ in order to find the normal bow-tie solution. Next we introduce the new variable $\tau = t^2/2$ and one more function $b'(t)$ such that $b_1(t) = tb'(t)$. Then the evolution equations take the form

$$ib' + 2i\tau\tfrac{db'}{d\tau} = \tfrac{1}{2}\epsilon b_2 + \sqrt{2}\sum_k v_k a_k$$

$$i\tfrac{db_2}{d\tau} = \tfrac{1}{2}\epsilon b' \tag{2.12}$$

$$i\tfrac{da_k}{d\tau} = \beta_k a_k + \sqrt{2}b', \quad (k \neq 0)$$

The letter set of equations contains multiplication over the independent variable τ only in a single equation. This means that the Fourier transformation reduces the problem to a single first order linear differential equation which is always solvable. However the proper choice of integration contour is rather complicated and further calculations are very tedious, although straightforward; therefore they are skipped in this text and only final results are shown. More details can be found in the original work [40]. Unlike the DOM the solution of the GBTM is not reducible to just application of transition probability formula for two states at every level intersection. However the independent crossing approximation that trivially takes into account the phase interference does reproduce all transition amplitudes in GBTM. The rules that can be used in evaluation of the transition amplitudes could be summarized as follows.

(i) Only forward propagation in time is considered.

(ii) The phase factors that are gained in the course of time propagation between crossings of adiabatic energy curves adjacent on the time axis should be set to zero.

(iii) The crossing of two diabatic potential curves induces rearrangement within the related two-dimensional subspace of Hilbert space. It is described by the transformation matrix

$$\begin{pmatrix} \sqrt{p_j} & i\sqrt{1-p_j} \\ i\sqrt{1-p_j} & \sqrt{p_j} \end{pmatrix} \tag{2.13}$$

where p_j is the Landau-Zener probability of diabatic passage of the related crossing, i.e. the one provided by naive application of the Landau-Zener formula for a corresponding level crossing of two states disregarding all others. The expression (2.13) shows that the dynamic phase $\frac{1}{2}\pi$ is gained in the transition from one diabatic state to another.

C. The Brundobler-Elser hypothesis and the absence of counterintuitive transitions

Although a few important classes of exactly solvable multistate models have been known for long time [32, 33] the interest toward the multistate Landau-Zener problem has grown up after the work of Brundobler and Elser [35] who noticed that for any model of the form (2.1) there are elements of the transition probability matrix that can be found by a simple application of the two state Landau-Zener formula at every intersection of diabatic energies. Particularly, they presented an empirical formula for the diagonal element of the scattering matrix for the state whose diabatic energy level has the highest slope i.e. if k is the index of the state with $\beta_k = \max(\beta_1 \dots \beta_N)$ or $\beta_k = \min(\beta_1 \dots \beta_N)$ then

$$|S_{kk}(+\infty, -\infty)| = exp\left(-\pi \sum_{i\,(i\neq k)} \frac{|A_{ki}|^2}{|\beta_k - \beta_i|}\right) \tag{2.14}$$

where A_{ij} are nondiagonal elements of the Hamiltonian (2.1). The formula (2.14) is confirmed by all known exactly solvable models with finite number of states [32, 33, 37, 38, 42, 28] and by multiple numerical checks. The authors of [35] speculated that this finding probably indicates that the whole problem (2.1) can be solved exactly or at least can be understood in terms of the two-level crossings. The work [70] demonstrated that (2.14) follows from a simple analytical continuation of the asymptotic solution into the complex time, although such a procedure fails to predict correctly other diagonal elements of the scattering matrix.

In the spirit of the Brundobler-Elser hypothesis Sinitsyn [43] conjectured that the independent crossing approximation can be exact for another set of scattering matrix elements in any multistate model of the type (2.1) that contains a band or bands of parallel levels (example of such a model is the Demkov-Osherov model, that includes a band of parallel levels).

Assume that instead of one state with the highest slope of diabatic energy level there is a band of an arbitrary number of states having the same highest slope so that diabatic energies in this band are different only by constant parameters ϵ_m. If we assume a "semiclassical" approximation where transition between any two states happen only at the corresponding crossing point of their diabatic energies then there are elements of the transition probability matrix that would be zero in this approximation. Such transitions are called counterintuitive. Thus in the model shown in Fig.4 transitions from the state 1 to states 2 and 3 and from the state 2 to the state 3 are counterintuitive.

Generally for the model (2.1), if $\beta_m = \beta_n = \max(\beta_1 \dots \beta_N)$ then the transition from the state m to the state n of the same band would be counterintuitive if $\epsilon_m < \epsilon_n$. Correspondingly, if $\beta_m = \beta_n = \min(\beta_1 \dots \beta_N)$ then the transition is counterintuitive if $\epsilon_m > \epsilon_n$. It is argued in [43], that in the multistate Landau-Zener model with linear

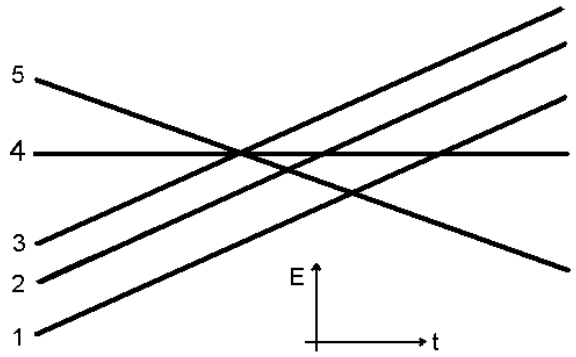

Fig. 4. Diabatic energies of a 5-state Landau-Zener model. The choice of parameters is as follows, $\beta_1 = \beta_2 = \beta_3 = 1$, $\beta_4 = 0$, $\beta_5 = -0.8$, $\epsilon_1 = 0$, $\epsilon_2 = 0.3$, $\epsilon_3 = 0.5$, $\epsilon_4 = 0$, $\epsilon_5 = 0.4$.

time-dependence of diabatic energies, such counterintuitive transitions have exactly zero probability, i.e. without assuming any semiclassical approximation for any model of the type (2.1), if the transition from the state m to the state n is counterintuitive, then

$$|S_{nm}(+\infty, -\infty)| = 0 \tag{2.15}$$

The rigorous mathematical proof of the Brundobler-Elser (2.14) conjecture and of the "no-go" conjecture (2.15) for counterintuitive transitions is still missing. However, these hypothesis can be understood by the approach similar to the one used by Landau in the two state calculations [1]. Since only asymptotic magnitudes of the amplitudes are needed, one can analytically extend the evolution (2.1) to imaginary time and choose the evolution path so that always $|t| \to \infty$. The distances between instantaneous eigenenergies of the Hamiltonian then remain always large, namely of the order of $|\beta_i - \beta_j|t >> A_{ij}$ for the states $i \neq j$ and hence one can try to use the adiabatic approximation

$$a_i(t) = e^{-i\int_{t_0}^{t} \epsilon_i(t)dt} a_i(t_0) \tag{2.16}$$

where the state a_i has the leading asymptotic $a_i \sim \exp(-i\beta_i t^2/2)$ at $t \to -\infty$.

The approximation (2.16) is valid generally only if there are no other states whose amplitudes become exponentially large in comparison with (2.16). Suppose that the state a_0 has the largest slope of adiabatic energy β_0 at $t \to -\infty$ and is initially occupied. In this case it is convenient to choose the time-path as shown in Fig.5 with $t = R\exp(i\phi)$ where $R \to \infty$ and ϕ decreases from π to zero. One can always change variables so that $\beta_0 = 0$ and $\beta_i < 0$ for states with slopes $\beta_i \neq \beta_0$ [35]. When ϕ changes in the interval from $3\pi/4$ to $\pi/4$, the amplitudes of states with slopes $\beta_i < 0$ are decreasing exponentially and become suppressed by the factor $\exp(C(\phi)\beta_i|t|^2/2)$ where $\beta_i < 0$ and $C(\phi)$ is a positive coefficient that depends only on the angle. We choose the asymptotics so that at the angle $\phi = 3\pi/4$ the state a_0 dominates over all others, i.e. is exponentially large in comparison to them. Then the states with $\beta_i < 0$ should not affect the adiabatic approximation in the interval $3\pi/4 > \phi > \pi/4$ since they can only decrease there. One can see that the condition that at $\phi = 3\pi/4$ the state a_0 is dominating also leads to the vanishing of the amplitudes of other states with $\beta_i < 0$ in the interval $\pi < \phi < 3\pi/4$ so that it is not forbidden to choose $|a_0(-\infty)| = 1$ and $|a_i(-\infty)| \to 0$, $(i \neq 0)$.

At the last part of the contour $\pi/4 > \phi > 0$ amplitudes of states with $\beta_i < 0$ grow from almost zero value but we know that at the end of the evolution they do not become larger than unity in absolute value. It means that they still remain small or comparable with a_0 at this interval and the formula (2.16) should be valid for the state a_0 during the whole evolution. Substituting the energy up to the first order correction in $1/|t|$

$$\epsilon_0(t) \sim \alpha_0 + \sum_i \frac{|A_{i0}|^2}{(\beta_0 - \beta_i)t} \tag{2.17}$$

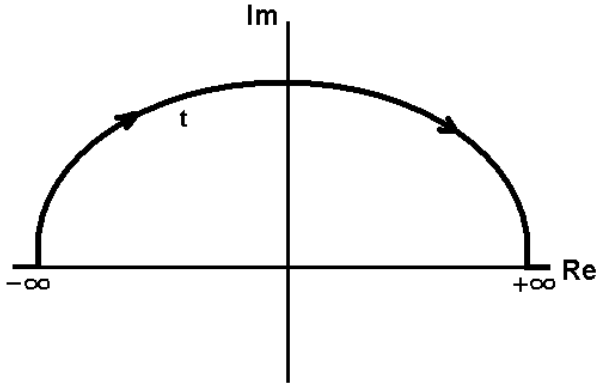

Fig. 5. The deformed time contour for the evolution from large negative to large positive times with $t = R\exp(i\phi)$, $R \to \infty$, $0 \leq \phi \leq \pi$.

into the formula for the transition probability

$$|S_{00}|^2 = \frac{|a_0(+\infty)|^2}{|a_0(-\infty)|^2} = exp\left(-2Im(\int\limits_C \epsilon_0(t)dt)\right) \tag{2.18}$$

one can find the Brundobler-Elser result (2.14). It is clear from this analysis why the formula (2.16) is generally not valid for other diagonal elements of the scattering matrix. If an initially filled state does not have the highest slope of the energy level there are states with higher slopes that grow exponentially and become large in the interval $3\pi/4 > \phi > \pi/4$ of the contour so that the adiabatic approximation becomes invalid in application to this state. To treat this case properly, one should investigate the Stokes phenomenon near all crossing points of diabatic energies [71] and still it remains unclear whether other diagonal elements of the S-matrix can be derived by similar approach.

This analysis becomes more complicated if there is more than one state having the same largest energy slope β_0. If such states have also a larger constant part of the diabatic energy $\epsilon_m > \epsilon_0$ they can grow in the first half of the contour as $\exp(C'(\phi)\epsilon_m|t|)$ i.e faster then the initially filled state a_0, but being initially vanishing,

amplitudes of such states can grow only due to transitions from the other states. At first half of the time-contour they are coupled only to states that are suppressed by much stronger exponents $\exp(C(\phi)\beta_i|t|^2/2)$, $(\beta_i < 0)$; therefore, we do not expect that they become large in comparison with a_0 up to $\phi = \pi/2$. In the second part of the path $\pi/2 < \phi < 0$ states with such an asymptotic $\exp(-i\epsilon_m t)$ already decrease exponentially and become suppressed in comparison with a_0; therefore we can expect that they do not break the approximation (2.16) for the state a_0 and have vanishing amplitudes at the end of the evolution. This is exactly in agreement with (2.15).

In addition to the above arguments, (2.15) is confirmed by all known exactly solvable classes featuring the possibility of counterintuitive transitions, namely by the Demkov-Osherov model [32], the generalized bow-tie model [38] and the model of two crossing bands of parallel levels [42]. Besides, a number of numerical simulations with arbitrary choices of parameters have been performed and all they supported the hypothesis (2.15). For example, in Fig.6 the time-dependence of the probabilities to find the system at states 2 and 3 in the model demonstrated schematically in Fig.4 are shown if initially only the state 1 was occupied. One can deduce that generally during the evolution these probabilities can be rather high (> 0.1) and show oscillating behavior, but asymptotically at $t \to +\infty$ they vanish. Numerically one can simulate the evolution only in a finite time interval. For the evolution from $t = -500$ to $t = 500$ and the same parameters as in Fig.6 numerical calculations predict $|S_{21}|^2 = 5.18 \times 10^{-7}$ and $|S_{31}|^2 = 3.11 \times 10^{-7}$. In comparison $|S_{11}|^2 = 0.234$, $|S_{41}|^2 = 0.295$ and $|S_{51}|^2 = 0.472$.

Although counterintuitive transitions have vanishing probabilities, the presence of the states 2 and 3 does affect other elements of the scattering matrix. Thus if we set all couplings of states 2 and 3 with all other states to zero then numerically calculated nondiagonal transition probabilities are $|S_{41}|^2 = 0.672$ and $|S_{51}|^2 = 0.094$

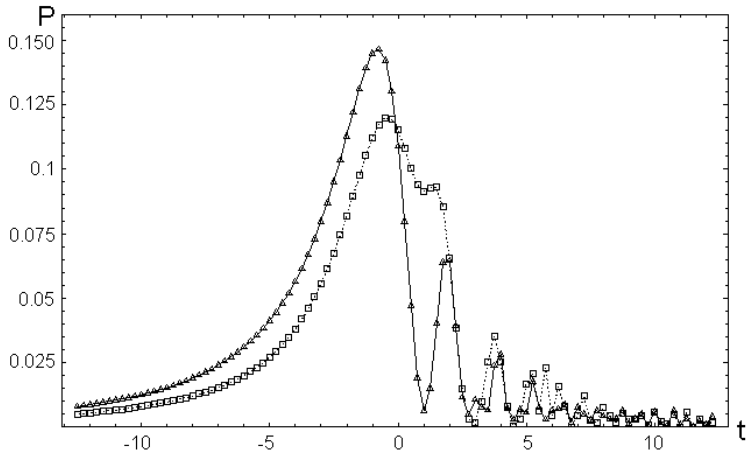

Fig. 6. Time dependence of the counterintuitive transition probabilities for the model in Fig.4. Triangles correspond to $P(t) = |S_{21}(t, -\infty)|^2$ and boxes show $P(t) = |S_{31}(t, -\infty)|^2$ The choice of nondiagonal elements of the Hamiltonian is $H_{12} = H_{13} = H_{23} = 0$, $H_{34} = 0.8$, $H_{35} = 0.3 + 0.24i$, $H_{24} = 0.1 + 0.7i$, $H_{25} = 0.5 + 0.1i$, $H_{14} = 0.4 + 0.12i$, $H_{15} = 0.25 + 0.2i$, $H_{45} = 0.6 + 0.9i$. The other elements are obtained by employing Hermitian properties of the matrix H.

that is different from the above numerical result.

D. Spin S Landau-Zener transitions

Consider a system with the total spin $S > 1/2$ in constant magnetic field H_x along x-direction and varying with time, much larger in average field $H_z(t)$ along z-direction. Its time evolution is regulated by the Hamiltonian:

$$\hat{H}_S = -h_x \hat{S}_x - h_z(t)\hat{S}_z \tag{2.19}$$

where $h_\alpha = g\mu_B H_\alpha$; $\alpha = x, z$ and $\hat{S}_z, \hat{S}_x$ are the spin operators. Such a spin evolution is realized when a Rydberg atom moves through a static magnetic or electric fields [72, 73, 74]. In the vicinity of its node $h_z(t)$ can be approximated by a linear function $h_z(t) = \dot{h}_z t$ where $\dot{h}_z$ is the time derivative of $h_z(t)$ taken at the node. After a proper

rescaling of time and energy the Hamiltonian (2.19) takes a following form:

$$\hat{H} = 2\gamma\hat{S}_x + t\hat{S}_z \tag{2.20}$$

It depends on one dimensionless parameter $\gamma = \frac{h_x}{\sqrt{\dot{h}_z}}$ (the LZ parameter). This Hamilton operator belongs to the $SU(2)$ algebra. The evolution of an arbitrary spin S in a varying magnetic field can be derived from the solution of Schrödinger equation for spin 1/2 in the same field [75]. The corresponding evolution operator is an operator of rotation belonging to the group $SU(2)$ and acting in its irreducible representation. Since the composition law does not depend on a specific representation, the resulting evolution operator represents the same rotation for any spin. Thus, the problem is reduced to the expression of the rotation operator for spin S if it is known for a spin 1/2.

The multi-spinor technique is most appropriate for this purpose (see, for example [75], ch. VIII). In general the spin S state can be represented as a direct symmetric product of 2S spin 1/2 states:

$$|S,m\rangle = \sqrt{\frac{(S+m)!(S-m)!}{(2S)!}}\left(|++...+--...-\rangle + |++...-+-...-\rangle + ...\right) \tag{2.21}$$

where each ket contains $S+m$ spins up and $S-m$ spins down and all permutations are performed. Let the $SU(2)$ matrix rotating spin 1/2 states is:

$$u = \begin{pmatrix} a & b \\ -b^* & a^* \end{pmatrix} \tag{2.22}$$

with $|a|^2 + |b|^2 = 1$. Equivalently an individual spinor is transformed according to:

$$
\begin{aligned}
|+\rangle &\rightarrow \quad a\,|+\rangle + b\,|-\rangle\,; \\
|-\rangle &\rightarrow -b^*\,|+\rangle + a^*\,|-\rangle
\end{aligned}
\tag{2.23}
$$

The transformation for the state (2.21) can be obtained as the direct product of transformations (2.23):

$$
\begin{aligned}
|S,m\rangle \rightarrow \sqrt{\frac{(S+m)!(S-m)!}{(2S)!}}\Big[a^{S+m}(-b^*)^{S-m}\,|S,S\rangle + \\
\sqrt{2S}\left(\frac{(2S-1)!}{(S-m-1)!(S+m)!}a^{S+m}(-b^*)^{S-m-1}a^* + \right. \\
\left.\frac{(2S-1)!}{(S+m-1)!(S-m)!}a^{S+m-1}(-b^*)^{S-m}b\right)|S,S-1\rangle\,...\Big]
\end{aligned}
\tag{2.24}
$$

A general matrix element for the spin S irreducible representation of the rotation operator $\langle m \mid U_S \mid m'\rangle$ is expressed in terms of a, b, a^*, b^* in the following way:

$$
\langle m \mid U_S \mid m'\rangle = \left[\frac{(S+m')!(S-m')!}{(S+m)!(S-m)!}\right]^{1/2} a^{m'+m}b^{m'-m}P_{S-m'}^{m'-m,m'+m}(2|a|^2-1) \tag{2.25}
$$

where $P_n^{a,b}(x)$ are the Jacobi polynomials [31]. The matrix elements possess following symmetry properties: $\langle -m \mid U_S \mid -m'\rangle = (-1)^{|m|+|m'|}\langle m \mid U_S \mid m'\rangle^*$, $|\langle m \mid U_S \mid m'\rangle| = |\langle m' \mid U_S \mid m\rangle| = |\langle -m' \mid U_S \mid -m\rangle|$.

Equation (2.25) displays oscillations of matrix elements associated with oscillatory behavior of the Jacobi polynomials. The number of nodes $N(m,m')$ of the matrix elements $\langle m \mid U_S \mid m'\rangle$ can be determined geometrically as the number of the square shell to which it belongs in the square matrix. We accept the number of the external square with $max(|m|,|m'|) = S$ for zero and number is increasing when the square shell size is decreasing. Analytically $N(m,m') = S - max(|m|,|m'|)$. Due to symmetry several matrix elements (2, 4 or 8) become zero at the same value of $|a|$

or γ. Central matrix elements have maximal number of nodes (S for integer spins, $S-1/2$ for half-integer spins).

The scattering matrix for our problem can be found from the general expression (2.25) by substitution:

$$a=\exp(-\pi\gamma^2),\quad b=-\frac{\sqrt{2\pi}\exp\left(\frac{\pi\gamma^2}{2}+\frac{i\pi}{4}\right)}{\gamma\Gamma(-i\gamma^2)} \tag{2.26}$$

which follows from comparison of equations (1.3), (2.22). To make our results more visible, we present the scattering matrices for spins $S=1$ and $S=3/2$.

$S=1$

$$U_1=\begin{pmatrix} a^2 & -\sqrt{2}ab^* & b^{*2} \\ \sqrt{2}ab & |a|^2-|b|^2 & -\sqrt{2}a^*b^* \\ b^2 & \sqrt{2}a^*b & a^{*2} \end{pmatrix} \tag{2.27}$$

$S=3/2$

$$U_{3/2}=\begin{pmatrix} a^3 & -\sqrt{3}a^2b^* & \sqrt{3}ab^{*2} & -b^{*3} \\ \sqrt{3}a^2b & (|a|^2-2|b|^2)a & (|b|^2-2|a|^2)b^* & \sqrt{3}a^*b^{*2} \\ \sqrt{3}ab^2 & (2|a|^2-|b|^2)b & (|a|^2-2|b|^2)a^* & -\sqrt{3}a^{*2}b^* \\ b^3 & \sqrt{3}a^*b^2 & \sqrt{3}a^{*2}b & a^{*3} \end{pmatrix} \tag{2.28}$$

Additionally the matrix of probabilities for $S=1$:

$$W=\begin{pmatrix} e^{-4\pi\gamma^2} & 2(e^{-2\pi\gamma^2}-e^{-4\pi\gamma^2}) & \left(1-e^{-2\pi\gamma^2}\right)^2 \\ 2(e^{-2\pi\gamma^2}-e^{-4\pi\gamma^2}) & \left(1-2e^{-2\pi\gamma^2}\right)^2 & 2(e^{-2\pi\gamma^2}-e^{-4\pi\gamma^2}) \\ \left(1-e^{-2\pi\gamma^2}\right)^2 & 2(e^{-2\pi\gamma^2}-e^{-4\pi\gamma^2}) & e^{-4\pi\gamma^2} \end{pmatrix} \tag{2.29}$$

As a consequence of Jacobi polynomials oscillations, the matrix elements of inner

squares have nodes at some special values of γ. Thus, the element $(U_1)_{00}$ is zero at $|a|^2 = 1/2$ or at $\gamma = \sqrt{\frac{1}{2\pi}\ln 2} \approx 0.332$. Elements of the matrix $(U_{3/2})_{1/2,1/2} = (U_{3/2})^*_{-1/2,-1/2}$ become zero at $|a|^2 = 2/3$, i.e. at $\gamma = \sqrt{\frac{1}{2\pi}\ln\frac{3}{2}} \approx 0.254$. Other two matrix elements $(U_{3/2})_{1/2,-1/2} = (U_{3/2})^*_{-1/2,1/2}$ become zero at $|a|^2 = 1/3$, i.e. at $\gamma = \sqrt{\frac{1}{2\pi}\ln 3} \approx 0.418$. In the matrix U_2 4 independent matrix elements have nodes: $(U_2)_{11}$ at $|a|^2 = 3/4$; $(U_2)_{00} = 0$ at $|a|^2 = \frac{1}{2}\left(1 \pm \frac{1}{\sqrt{3}}\right)$; $(U_2)_{10} = 0$ at $|a|^2 = 1/2$; $(U_2)_{1,-1} = 0$ at $|a|^2 = 1/4$.

E. The multiparticle Landau-Zener model

Ideas in the background of the arbitrary spin solution can be generalized so that any solvable model generates a class of different solvable multistate Hamiltonians [28]. The main trick is to suppose that the evolution equations for some solvable multistate model are not Schrödinger but rather Heisenberg equations i.e. they are written not for amplitudes but rather for some operators. Since equations are still linear in operators this problem is solvable too. However to make a physical meaning of this evolution one should find the Hamiltonian that generates such operator equations in Heisenberg representation. This Hamiltonian should belong to the same algebra as operators that participate in the evolution equations. Still there is an arbitrariness in choosing the Hilbert space. If such a Hamiltonian is found and if it keeps invariant some finite-dimensional space of quantum states, in that subspace it can have a matrix form which belongs to the class of multistate Landau-Zener models. Since the evolution equations for operators are solvable the corresponding evolution equations for amplitudes of such a generated multistate Landau-Zener model are solvable too.

Besides the interest in new solutions, the idea to consider the Landau-Zener evolution for operators rather than for amplitudes can be fruitful also in problems

with level crossings when more than one particle participate in the evolution. In this subsection the generalizations of the Landau-Zener models to Heisenberg evolution of Bose and Fermi operators are applied to generate new solvable multistate Landau-Zener models and applications of such equations in quantum dots and in Bose condensates are demonstrated.

1. The bosonic Landau-Zener model

Consider a Hamiltonian that describes the interaction of four bosonic fields $\hat{a}$, $\hat{b}$, $\hat{c}$, $\hat{d}$:

$$\begin{aligned}\hat{H} = (\beta_1 t + E_1)\hat{a}^\dagger\hat{a} + (\beta_3 t + E_3)\hat{d}^\dagger\hat{d} + (\beta_2 t + E_2)\hat{c}^\dagger\hat{c} + (\beta_4 t + E_4)\hat{b}^\dagger\hat{b} + \\ g(\hat{a}^\dagger\hat{b} + \hat{b}^\dagger\hat{a}) + \gamma(\hat{c}^\dagger\hat{d} + \hat{d}^\dagger\hat{c})\end{aligned} \tag{2.30}$$

This Hamiltonian depends explicitly on time and conserves the total number of particles in the system. Therefore it can be considered independently in subspaces with fixed total number of particles. Let $|0\rangle$ be the vacuum state. The Hamiltonian (2.30) describes the evolution of two disjointed systems. However, being projected onto the 2-particle sector, its matrix form looks less trivial. The complete two-particle sector is the 10-dimensional Hilbert space spanned onto direct products of any two single-particle states. The four-dimensional subspace R_4 of the 2-particle sector spanned onto vectors:

$$\begin{aligned}|1\rangle &= \hat{a}^\dagger\hat{c}^\dagger|0\rangle \\ |2\rangle &= \hat{a}^\dagger\hat{d}^\dagger|0\rangle \\ |3\rangle &= \hat{d}^\dagger\hat{b}^\dagger|0\rangle \\ |4\rangle &= \hat{c}^\dagger\hat{b}^\dagger|0\rangle\end{aligned} \tag{2.31}$$

is invariant with respect to the action of the Hamiltonian (2.30). Hence, if the initial state belongs to this subspace, the state vector at any time remains in R_4:

$$|\psi(t)\rangle = c_1(t)|1\rangle + c_2(t)|2\rangle + c_3(t)|3\rangle + c_4(t)|4\rangle \tag{2.32}$$

In the basis (2.31) the Hamiltonian (2.30) has the following 4x4 matrix form:

$$H = \begin{pmatrix} (\beta_1+\beta_2)t + (E_1+E_2) & \gamma & 0 & g \\ \gamma & (\beta_1+\beta_3)t + (E_1+E_3) & g & 0 \\ 0 & g & (\beta_3+\beta_4)t + (E_3+E_4) & \gamma \\ g & 0 & \gamma & (\beta_2+\beta_4)t + (E_4+E_2) \end{pmatrix} \tag{2.33}$$

The problem described by the Hamiltonian (2.33) belongs to the multistate Landau-Zener class (2.1).

In the Heisenberg representation the evolution equations decouple into two pairs of equations for bosonic operators:

$$\begin{aligned} i\dot{\hat{a}} &= (\beta_1 t + E_1)\hat{a} + g\hat{b} \\ i\dot{\hat{b}} &= (\beta_4 t + E_4)\hat{b} + g\hat{a} \end{aligned} \tag{2.34}$$

and

$$\begin{aligned} i\dot{\hat{c}} &= (\beta_2 t + E_2)\hat{c} + \gamma\hat{d} \\ i\dot{\hat{d}} &= (\beta_3 t + E_3)\hat{d} + \gamma\hat{c} \end{aligned} \tag{2.35}$$

Let $\hat{a}_0, \hat{b}_0, \hat{c}_0, \hat{d}_0$ denote the operators $\hat{a}, \hat{b}, \hat{c}, \hat{d}$ at the initial moment of evolution. Then the solutions of equations (2.34) and (2.35) are:

$$\begin{aligned} \hat{a}(t) &= S_{11}(t)\hat{a}_0 + S_{12}(t)\hat{b}_0 \\ \hat{b}(t) &= S_{21}(t)\hat{a}_0 + S_{22}(t)\hat{b}_0 \end{aligned} \tag{2.36}$$

$$\begin{aligned} \hat{c}(t) &= S'_{11}(t)\hat{c}_0 + S'_{12}(t)\hat{d}_0 \\ \hat{d}(t) &= S'_{21}(t)\hat{c}_0 + S'_{22}(t)\hat{d}_0 \end{aligned} \tag{2.37}$$

Here S_{ij} and S'_{ij} are the matrix elements of the evolution operators for (2.34) and (2.35), respectively. Due to the linearity they are the same for the operator and numerical functions obeying these differential equations. Hence, we can extract them directly from the solution of the two-state LZ problem. For the evolution from $t = -\infty$ to $t = +\infty$ their squares of modulus are:

$$\begin{aligned} p_1 &\equiv |S_{11}|^2 = |S_{22}|^2 = e^{-2\pi g^2/|\beta_1-\beta_4|} \\ q_1 &\equiv |S_{12}|^2 = |S_{21}|^2 = 1 - p_1 \\ p_2 &\equiv |S'_{11}|^2 = |S'_{22}|^2 = e^{-2\pi \gamma^2/|\beta_2-\beta_3|} \\ q_2 &\equiv |S'_{12}|^2 = |S'_{21}|^2 = 1 - p_2 \end{aligned} \tag{2.38}$$

Returning to the four-state LZ problem in the two-particle sector considered earlier, each state $|\gamma\rangle$ of this subspace is the direct product of states from two independent subspaces of the one-particle sector $|j\rangle = |\alpha_j\rangle \bigotimes |\mu_j\rangle$, $\alpha_j = 1, 2; \mu_j = 3, 4$ (here 1,2,3,4 enumerate single-particle state, for example $|1\rangle = a^+|0\rangle$). The evolution matrix is also the direct product of evolution matrices in the independent subspaces of the one-particle sectors: $U(t) = U_\alpha(t) \bigotimes U_\mu(t)$. Therefore transition matrix elements and probabilities P_{ij} in the considered subspace are factorized:

$$P_{ij} = p_{\alpha_i \alpha_j} p_{\mu_i \mu_j} \tag{2.39}$$

In terms of the LZ probabilities for two-level problems introduced earlier the transition probability matrix P, whose elements are defined by equation (2.39), reads:

$$P = \begin{pmatrix} p_1p_2 & p_1q_2 & q_1q_2 & p_1q_2 \\ p_1q_2 & p_1p_2 & q_1p_2 & q_1q_2 \\ q_1q_2 & p_2q_1 & p_1p_2 & p_1q_2 \\ q_1p_2 & q_1q_2 & p_1q_2 & p_1p_2 \end{pmatrix} \tag{2.40}$$

This result does not depend on the parameters E_i. Scattering matrices $S_{ij}(t)$ and $S'_{ij}(t)$ are known for any t [2] which makes it possible to find the evolution operator at any time in the Schrödinger representation.

The above example looks trivial since the generated model is just a factorization of two independent simpler models. However this is not always just a factorization. For example the model of LZ transitions in arbitrary spin, that is reviewed in the previous section, can be derived from the following bosonic Hamiltonian:

$$\hat{H} = t\hat{a}^\dagger\hat{a} - t\hat{b}^\dagger\hat{b} + g(\hat{a}^\dagger\hat{b} + \hat{b}^\dagger\hat{a}) \tag{2.41}$$

In the single-particle sector the Hamiltonian (2.41) leads to a simple two-state LZ model. In the N-boson sector the Schrödinger equation for diabatic states coincides with ones for a spin $S = N/2$ in magnetic fields. This construction is an application of the Schwinger bosons [76] to the LZ problem. By solving trivial equations for Schwinger operators, one can derive the solution for LZ transitions in any spin.

2. The fermionic Landau-Zener model and a charge transport problem

In similar fashion to the bosonic problem, the fermionic systems can lead to Heisenberg equations for annihilation operators that have the same structure as Shrödinger equations for amplitudes for some exactly solvable multistate Landau-Zener model. In the Shrödinger representation such a Fermi system with fixed number of particles is equivalent to a new solvable multistate Landau-Zener model.

Consider a quantum dot coupled to an external reservoir like the system shown in Fig.7 so that initially some of the reservoir energy levels are filled with electrons, the others are empty. Assume the dot has only one electron bound state in the energy region of interest. The energy of this state in real semiconductors can be regulated by the gate voltage; therefore the variation of the gate voltage with time generates

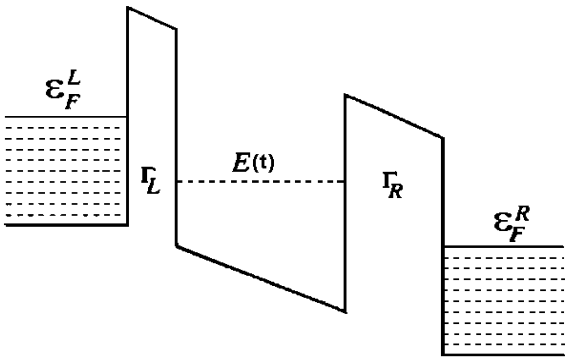

Fig. 7. A single energy level in a potential well coupled to two leads at zero temperature. Electron states in leads are filled up to Fermi energies, that can be different in right and left leads.

time dependence of the dot's electronic level.

The simplest possible choice of the Hamiltonian of electrons in the dot and the reservoir reads:

$$H = \sum_{n=1}^{N} E_n \hat{c}_n^\dagger \hat{c}_n + E(t)\hat{c}_0^\dagger \hat{c}_0 + \sum_n g_n(\hat{c}_n^\dagger \hat{c}_0 + \hat{c}_0^\dagger \hat{c}_n) \tag{2.42}$$

Here $\hat{c}_0$ is the fermionic operator that annihilates the electron on the dot level and $\hat{c}_n$ is the annihilation operator for the level E_n of the reservoir; $E(t)$ is the time-dependent energy of the dot state. The last term in (2.42) describes the tunneling between the leads and the single level in the quantum dot. All interactions among electrons are ignored except the one due to Pauli principle.

Similar time-dependent single-particle problems for quantum dots have been already considered in [13] although the possibility of the exact solution in the multi-particle sector was omitted.

In the context of the LZ theory, the time-dependence of the dot energy can be approximated by a linear function: $E(t) = \beta t$. The Heisenberg operator equations

corresponding to Hamiltonian (2.42) are:

$$\begin{aligned} i\dot{\hat{c}}_0 &= \beta t \hat{c}_0 + \sum_n g_n \hat{c}_n \\ i\dot{\hat{c}}_n &= E_n \hat{c}_n + g_n \hat{c}_0 \end{aligned} \tag{2.43}$$

Due to the linear structure of these equations the solution can be formally written in the matrix form:

$$\hat{\mathbf{c}}(t) = \hat{S}(t, t_0)\ \hat{\mathbf{c}}(t_0) \tag{2.44}$$

where $\hat{\mathbf{c}} = (\hat{c}_0, \hat{c}_1, \ldots, \hat{c}_n)$

As in the previous section, the evolution matrix $\hat{S}(t, t_0)$ is completely determined by the coefficients of the differential equations (2.43) and is the same for operator and c-function solutions. Hence, it is enough to solve (2.43) with all operators replaced by c-functions. Such a system of equations coincides with that of the Demkov-Osherov model [32].

The probabilities to find an electron on a particular n-th level after all intersections are.

$$\begin{aligned} P_n &= < \hat{c}_n^\dagger(t \to +\infty)\hat{c}_n(t \to +\infty) > = \textstyle\sum_{n_1}\sum_{n_2} S^*_{nn_1} S_{nn_2} < \hat{c}^\dagger_{n_1}(t \to -\infty)\hat{c}_{n_2}(t \to -\infty) > = \\ &= \sum_{n_f} |S_{n,n_f}|^2 \end{aligned} \tag{2.45}$$

where $S_{ij} = S_{ij}(t \to +\infty)$, $(t_0 \to -\infty)$ and the summation in the final expression is taken over the initially filled states only.

The scattering matrix elements S_{n,n_f} are given in (2.8). If the band of electron states in the external system is continuous then it is reasonable to use the approximation, in which $g(E) = g_n \to 0$ while the value $\Gamma(E) = 2\pi\rho(E)|g(E)|^2$ is kept finite. Here $\rho(E)$ is the density of states in the band and the elements of scattering matrix become $|S_{0l}|^2 = \frac{2\pi g_l^2}{\beta} \exp \int\limits_{E_l}^{E_n} \frac{-\Gamma(E)}{\beta} dE$

Consider a dot that is connected to two leads. The left lead is characterized by the coupling function $g_L(E)$ and the densities $\rho_L^f(E)$, $\rho_L^e(E)$ where f and e refer to the filled and empty states in the left lead ($\rho_L(E) = \rho_L^f(E) + \rho_L^e(E)$), analogously we can define the quantities $g_R(E)$, $\rho_R^f(E)$, $\rho_R^e(E)$ for the right lead. Moreover it is more convenient to introduce the following notations: $\Gamma_L^f(E) = 2\pi\rho_L^f(E)|g_L(E)|^2$, $\Gamma_L^e(E) = 2\pi\rho_L^e(E)|g_L(E)|^2$, $\Gamma_R^f(E) = 2\pi\rho_R^f(E)|g_R(E)|^2$, $\Gamma_R^e(E) = 2\pi\rho_R^e(E)|g_R(E)|^2$, $\Gamma^f = \Gamma_R^f + \Gamma_L^f$, and $\Gamma^e = \Gamma_R^e + \Gamma_L^e$.

If the dot state was initially empty and if this state crosses the region from energy E_1 to energy E_2, then the continuous approximation (2.45) leads to the following probability for the dot level to be finally filled after all Landau-Zener transitions:

$$p_f(E_2) = P_0 = \int_{E_1}^{E_2} \frac{\Gamma^f(E')}{\beta} \exp\left[-\frac{1}{\beta}\int_{E'}^{E_2}(\Gamma^f(E) + \Gamma^e(E))dE\right]dE' \tag{2.46}$$

If the dot level was initially filled, it is necessary to add $|S_{00}|^2 = e^{-\frac{1}{\beta}\int_{E_1}^{E_2}(\Gamma^f(E)+\Gamma^e(E))dE}$ to (2.46). One can check that the result (2.46) is the solution of the following system of differential equations:

$$\begin{aligned} \beta\tfrac{dp_f(E)}{dE} &= -\Gamma^e(E)p_f(E) + \Gamma^f(E)p_e(E) \\ \beta\tfrac{dp_e(E)}{dE} &= -\Gamma^f(E)p_e(E) + \Gamma^e(E)p_f(E) \end{aligned} \tag{2.47}$$

here $p_e(E) = 1 - p_f(E)$ is the probability that the dot level will be empty when it has energy E. The equation for the charge that is transferred to the right lead can be derived in a similar way

$$\frac{dQ(E)}{dE} = (e/\beta)(\Gamma_R^e(E)p_f(E) - \Gamma_R^f(E)p_e(E)) \tag{2.48}$$

With this result one can calculate the total charge transferred through the dot from

the left lead to the right lead at zero temperature and a fixed bias that leads to a difference of Fermi energies in the left and in the right leads. Assume that the dot level was initially much lower than both Fermi levels and it was filled. Then the energy of this state grows linearly with time crossing both Fermi levels during the evolution. Since transitions will proceed presumably when the dot level is between Fermi energies of the leads, one can apply the following approximations: $\Gamma_L^f(E) = \Gamma_L(1-\theta(E-\epsilon_F^L))$, $\Gamma_L^e(E) = \Gamma_L\theta(E-\epsilon_F^L)$, $\Gamma_R^f(E) = \Gamma_R(1-\theta(E-\epsilon_F^R))$ and $\Gamma_R^e(E) = \Gamma_R\theta(E-\epsilon_F^L)$ with Γ_R and Γ_L are constant. To find the total charge that is transferred to the right lead one can formally put the final dot state energy equal to infinity in the solution of the equations (2.47) and (2.48). In the result the total charge transfered to the right lead is

$$Q = e\left[\frac{\Gamma_R\Gamma_L}{(\Gamma_R+\Gamma_L)}(\frac{\epsilon_F^L-\epsilon_F^R}{\beta}) + \frac{\Gamma_R}{(\Gamma_R+\Gamma_L)}\right] \tag{2.49}$$

At $\epsilon_F^L = \epsilon_F^R$ we find $Q = e\Gamma_R/(\Gamma_R+\Gamma_L)$, which can be interpreted as the electron charge e multiplied by the probability for the electron that is initially placed into the dot to transfer to the right lead.

3. A solvable model of bands crossing

The Hamiltonian (2.42) projected onto the k-particle sector generates the evolution in the Hilbert space of dimensionality $(N+1)!/(k!(N+1-k)!)$. If the single-particle Hamiltonian laying in the background is the same as that of the Demkov-Osherov model, then all such models are reducible to this single-particle one.

Generalized Landau-Zener models that deal with intersections of bands of parallel levels are important in many applications such as in tunneling of nanomagnets coupled to nuclear spins [8] and in charge transport in quantum dots [42].

Up to now only two exact solutions of this type were known: Demkov-Osherov solution and the case of the infinite number of states in bands that equally interact with states of another band [36]. For an important case of a finite number of states in bands that is not equal to unity, exact solutions for all transition probabilities have not been found yet though the absence of counterintuitive transitions was analytically proved [42]. Nearly-exact solution valid in the quasidegeneracy approximation was found and investigated in [15]. The method of extending the Landau-Zener evolution to the operators can be employed to generate exactly solvable models with interband transitions.

Consider a system with the Hamiltonian of four interacting Fermi operators.

$$H = E_1\hat{b}^\dagger\hat{b} + E_2\hat{c}^\dagger\hat{c} + t\hat{d}^\dagger\hat{d} + g_1(\hat{a}^\dagger\hat{d} + \hat{d}^\dagger\hat{a}) + g_2(\hat{b}^\dagger\hat{d} + \hat{d}^\dagger\hat{b}) + g_3(\hat{c}^\dagger\hat{d} + \hat{d}^\dagger\hat{c}) \qquad (2.50)$$

Let $E_2 > E_1 > 0$. The solution of the operator evolution equation can be written in the form:

$$\begin{pmatrix} \hat{d}(t) \\ \hat{a}(t) \\ \hat{b}(t) \\ \hat{c}(t) \end{pmatrix} = S(t,t_0) \begin{pmatrix} \hat{d}(t_0) \\ \hat{a}(t_0) \\ \hat{b}(t_0) \\ \hat{c}(t_0) \end{pmatrix}, \qquad (2.51)$$

where $S(t,t_0)$ is the matrix of evolution for a 4-state Demkov-Osherov model. Lets restrict the Hilbert space to the subspace of only two particles. It includes six states: $|1\rangle = \hat{d}^\dagger\hat{a}^\dagger|0\rangle$, $|2\rangle = \hat{d}^\dagger\hat{b}^\dagger|0\rangle$, $|3\rangle = \hat{d}^\dagger\hat{c}^\dagger|0\rangle$, $|4\rangle = \hat{a}^\dagger\hat{b}^\dagger|0\rangle$, $|5\rangle = \hat{a}^\dagger\hat{c}^\dagger|0\rangle$ and $|6\rangle = \hat{b}^\dagger\hat{c}^\dagger|0\rangle$. Similarly to the bosonic case, this subspace is invariant during the evolution process.

The Hamiltonian restricted to this subspace has the following matrix form:

$$H = \begin{pmatrix} t & 0 & 0 & -g_2 & -g_3 & 0 \\ 0 & t+E_1 & 0 & g_1 & 0 & -g_3 \\ 0 & 0 & t+E_2 & 0 & g_1 & g_2 \\ -g_2 & g_1 & 0 & E_1 & 0 & 0 \\ -g_3 & 0 & g_1 & 0 & E_2 & 0 \\ 0 & -g_3 & g_2 & 0 & 0 & E_1+E_2 \end{pmatrix} \tag{2.52}$$

Lets P_{ij} $(i,j = 1,...,6)$ be the probability to transit from the state j to the state i after the band crossing. The transition probabilities can be expressed in terms of the fermi-operators in the Heisenberg representation at $t \to \infty$.

$$\begin{aligned} P_{1n} &= \langle n|\hat{a}^\dagger \hat{a}\hat{d}^\dagger \hat{d}|n\rangle \quad P_{4n} = \langle n|\hat{a}^\dagger \hat{a}\hat{b}^\dagger \hat{b}|n\rangle \\ P_{2n} &= \langle n|\hat{b}^\dagger \hat{b}\hat{d}^\dagger \hat{d}|n\rangle \quad P_{5n} = \langle n|\hat{a}^\dagger \hat{a}\hat{c}^\dagger \hat{c}|n\rangle \\ P_{3n} &= \langle n|\hat{c}^\dagger \hat{c}\hat{d}^\dagger \hat{d}|n\rangle \quad P_{6n} = \langle n|\hat{b}^\dagger \hat{b}\hat{c}^\dagger \hat{c}|n\rangle \end{aligned} \tag{2.53}$$

Substituting (2.51) into (2.53) and employing the elements of the evolution matrix from (2.8) one can arrive to the following result:

$$P = \begin{pmatrix} p_2p_3 & q_1q_2p_3 & q_1q_3 & p_1q_2p_3 & p_1q_3 & 0 \\ 0 & p_1p_3 & p_1q_2q_3 & q_1p_3 & q_1q_2q_3 & p_2q_3 \\ 0 & 0 & p_1p_2 & 0 & q_1p_2 & q_2 \\ q_2 & q_1p_2 & 0 & p_1p_2 & 0 & 0 \\ p_2q_3 & q_1q_2q_3 & q_1p_3 & p_1q_2q_3 & p_1p_3 & 0 \\ 0 & p_1q_3 & q_2p_3p_1 & q_1q_3 & q_1q_2p_3 & p_2p_3 \end{pmatrix} \tag{2.54}$$

where

$$
\begin{aligned}
p_i &= e^{-2\pi|g_i|^2} \\
q_i &= 1 - p_i, \quad (i = 1, 2, 3)
\end{aligned}
\tag{2.55}
$$

F. Nonunitary Landau-Zener problem and dissociation of molecular Bose condensate

Similarly to fermionic Landau-Zener model, the bosonic one also may have applications on its own without just being useful in understanding the multistate Landau-Zener problem. Mathematically, the model in this section, when considered in the Hilbert space, corresponds to a multistate Landau-Zener model with infinite number of states. However its treatment is strongly simplified when writing its Hamiltonian in a secondary quantized form. The resulting evolution equations for operators only resemble the Landau-Zener evolution but are not exactly the same. Moreover if such equations were written for amplitudes, not for operators, such evolution would be nonunitary. Nevertheless the model is exactly solvable which allows to make quantitative predictions about corresponding physical system.

Imagine the process of molecular condensate dissociation such as $A_2 \rightarrow A + A$. It was studied for example in [63],[77]. The authors demonstrated that Bose condensate of single atoms [78] can be in squeezed state after molecular condensate dissociation. If chemical potentials of condensates are time dependent, the number of atoms and the squeezing parameters in the case of time dependent chemical potentials depend on the rate of energy curve crossing and on the coupling between the molecular and the atomic fields. If the initial atomic condensate exists in the system, then the result of the above reaction depends not only on the total number of atoms but also on initial phases of the molecular and the atomic condensates. Generally the single atoms after molecular dissociation should not be in the same quantum states (modes). The case of multimode condensate dynamics [79], [80], [81], [82] with time dependent

parameters is more complicated since it is described by the multistate Landau-Zener theory which generally does not provide an exact result for transition amplitudes. However, within reasonable approximations the problem can be reduced to the case of two atomic modes and one molecular mode interaction for which exact results are still possible since the equations for the operator evolution decouple.

The most general Hamiltonian that includes interactions and more than one atomic and molecular modes can be written as follows[82]:

$$\begin{aligned}\hat{H} &= \sum_{\mathbf{k}}(\mu_k(t)\hat{a}^\dagger_{\mathbf{k}}\hat{a}_{\mathbf{k}} + \nu_k(t)\frac{1}{2}\hat{\psi}^\dagger_{\mathbf{k}}\hat{\psi}_{\mathbf{k}}) + g_{k,k'}\sum_{\mathbf{k},\mathbf{k}'}\hat{\psi}^\dagger_{\mathbf{k}+\mathbf{k}'}\hat{a}_{\mathbf{k}}\hat{a}_{\mathbf{k}'} + \hat{\psi}_{\mathbf{k}+\mathbf{k}'}\hat{a}^\dagger_{\mathbf{k}}\hat{a}^\dagger_{\mathbf{k}'} \\ &+ \frac{1}{2}g_{int}\sum_{\mathbf{k},\mathbf{k}',\mathbf{k}''}\hat{a}^\dagger_{\mathbf{k}+\mathbf{k}'-\mathbf{k}''}\hat{a}^\dagger_{\mathbf{k}''}\hat{a}_{\mathbf{k}'}\hat{a}_{\mathbf{k}}\ ,\end{aligned}$$

here the chemical potentials μ_k and ν_k depend on time. The Landau-Zener model for multimode case becomes very complicated to solve even in the case when the molecular field is treated as a c-number (which is possible in the mean-field approximation with condition that the number of molecules is so large that it does not change strongly during the process). If the number of atoms in the atomic modes is much smaller than that of molecules, one can disregard the interaction term ($g_{int} = 0$). Suppose the system has only one highly populated mode of the molecular condensate. For example, molecular mode with total momentum equals zero. Due to momentum conservation atomic mode with momentum $\mathbf{k}$ would couple to atomic mode of momentum $-\mathbf{k}$ only. In such systems the problem is reduced to the interaction of two atomic modes with molecular condensate. Other molecular modes can also be considered but the effective coupling constants to those modes are proportional to the square root of the number of particles in them; therefore, one can disregard the interactions with the initially empty modes.

With such approximations the problem is reduced to finding dynamics of two atomic fields A and B interacting with one molecular field AB. Suppose $\hat{a}$, $\hat{b}$ and $\hat{\psi}$ are the annihilation operators of A, B and AB fields respectively. If one disregards the direct interaction between A and B during curve crossing then the Hamiltonian for molecular dissociation in the process $AB \rightarrow A + B$ is time dependent and can be written as follows:

$$\hat{H} = \mu_1(t)\hat{a}^\dagger\hat{a} + \mu_2(t)\hat{b}^\dagger\hat{b} + g\hat{\psi}^\dagger\hat{a}\hat{b} + g^*\hat{\psi}\hat{a}^\dagger\hat{b}^\dagger \tag{2.56}$$

where the molecular field energy is set to zero and $\hat{a}$ and $\hat{b}$ are annihilation operators of distinct atomic modes so $[\hat{a}^\dagger, \hat{b}] = 0$. If the number of molecules is large enough to suppose that it does not change considerably during the process one can substitute $\langle\hat{\psi}\rangle$ instead of $\hat{\psi}$. Substituting $g < \hat{\psi} >^* = \gamma$ into (2.56) the effective atomic Hamiltonian reads

$$\hat{H} = \mu_1(t)\hat{a}^\dagger\hat{a} + \mu_2(t)\hat{b}^\dagger\hat{b} + \gamma\hat{a}\hat{b} + \gamma^*\hat{a}^\dagger\hat{b}^\dagger \tag{2.57}$$

The model (2.57) but with identical atomic modes ($B = A$) was first considered in [63]. However generally atomic modes should be considered distinct and be described by distinct Bose operators. The Hamiltonian (2.57) with distinct A and B was considered by Kayali and Sinitsyn in [21] and also by Anglin in [22]. The Heisenberg equations for operators are

$$\begin{aligned} i\dot{\hat{a}} &= \mu_1(t)\hat{a} + \gamma^*\hat{b}^\dagger \\ i\dot{\hat{b}}^\dagger &= -\mu_2(t)\hat{b}^\dagger - \gamma\hat{a} \end{aligned} \tag{2.58}$$

and

$$\begin{aligned} i\dot{\hat{a}}^\dagger &= -\mu_1(t)\hat{a}^\dagger - \gamma\hat{b} \\ i\dot{\hat{b}} &= \mu_2(t)\hat{b} + \gamma^*\hat{a}^\dagger \end{aligned} \tag{2.59}$$

After the following change of variable

$$\begin{aligned} \hat{a} &\to \hat{a}e^{-i\int^t(\mu_1(t)/2-\mu_2(t)/2)dt} \\ \hat{b} &\to \hat{b}e^{i\int^t(\mu_1(t)/2-\mu_2(t)/2)dt} \\ \mu(t) &= \mu_1(t)/2+\mu_2(t)/2 \end{aligned} \tag{2.60}$$

equations (2.58),(2.59) read:

$$\begin{aligned} i\dot{\hat{a}} &= \mu(t)\hat{a}+\gamma^*\hat{b}^\dagger \\ i\dot{\hat{b}}^\dagger &= -\mu(t)\hat{b}^\dagger-\gamma\hat{a} \end{aligned} \tag{2.61}$$

and

$$\begin{aligned} i\dot{\hat{a}}^\dagger &= -\mu(t)\hat{a}^\dagger-\gamma\hat{b} \\ i\dot{\hat{b}} &= \mu(t)\hat{b}+\gamma^*\hat{a}^\dagger \end{aligned} \tag{2.62}$$

Molecular dissociation becomes substantial near the curve crossing points $\mu(t)=0$. According to the Landau-Zener theory, to estimate the transition amplitude one can approximate $\mu(t)$ linearly near the crossing points, hence the equations that should be considered are:

$$\begin{aligned} i\dot{\hat{a}} &= \beta t\hat{a}+\gamma^*\hat{b}^\dagger \\ i\dot{\hat{b}}^\dagger &= -\beta t\hat{b}^\dagger-\gamma\hat{a} \end{aligned} \tag{2.63}$$

The system (2.63) strongly reminds the Landau-Zener problem but it is not. The difference is in the additional minus sign in the second equation. If the system (2.63) were written for functions rather than operators, it would describe a nonunitary evolution. This is related to the fact that the number of particles in single atomic modes is not conserved. The corresponding second order differential equation for operator $\hat{a}$ reads:

$$\ddot{\hat{a}}+(\beta^2t^2-|\gamma|^2+i\beta^2)\hat{a}=0 \tag{2.64}$$

This equation belongs to the class of hypergeometric equations which are solvable and whose asymptotics are well known. The solution can be written in the form:

$$\hat{a}(t) = \phi_c(t)\hat{a}(t_0) + \phi_s(t)\hat{b}^\dagger(t_0) \tag{2.65}$$

with initial conditions:

$$\begin{aligned} \phi_c(t_0) = 1, \phi_s(t_0) = 0 \\ \dot{\phi}_c(t_0) = \beta t_0, \dot{\phi}_s(t_0) = -i\gamma^* \end{aligned} \tag{2.66}$$

The functions $\phi_c(t)$ and $\phi_s(t)$ are c-functions that are independent solutions of the equation (2.64). The equation (2.64) with the initial conditions (2.66) appears also in the case of dissociation into a single mode atomic condensate but with different magnitudes of parameters [63]. The average number of atoms in the A-mode is:

$$< \hat{a}^\dagger(t)\hat{a}(t) >= n_{st} + n_{sp} \tag{2.67}$$

where

$$n_{sp} = |\phi_s(t)|^2 \tag{2.68}$$

corresponds to the spontaneous transitions into the $A-$atomic vacuum state. This term does not appear if the problem is treated in the mean field approximation and is the result of quantum effects [63, 83, 84, 85]. The quantity n_{st} corresponds to the stimulated transitions in the case when atomic states were initially populated:

$$n_{st} = |\phi_c|^2 < \hat{a}^\dagger(t_0)\hat{a}(t_0) > +|\phi_s|^2 < \hat{b}^\dagger(t_0)\hat{b}(t_0) > +2Re(\phi_s^*\phi_c < \hat{a}(t_0)\hat{b}(t_0) >) \tag{2.69}$$

Solutions that satisfy our initial conditions are given by

$$\phi_c(t) = -i\gamma^* \frac{\phi_1^*(t_0)\phi_1(t) - \phi_2^*(t_0)\phi_2(t)}{W(\phi_1, \phi_2)} \tag{2.70}$$

$$\phi_s(t) = 2i\gamma^* \frac{\phi_2(t_0)\phi_1(t) - \phi_1(t_0)\phi_2(t)}{W(\phi_1, \phi_2)} \tag{2.71}$$

where $W(\phi_1, \phi_2) = i(2\beta/\gamma)exp(-\pi\lambda/2)$ and

$$\lambda = |\gamma|^2/(2\beta) \tag{2.72}$$

is the Landau-Zener parameter. The Functions ϕ_1 and ϕ_2 are two standard solutions of the parabolic cylinder equation with asymptotics at $t_0 \longrightarrow -\infty$:

$$\phi_1(t_0) \sim \frac{1}{\tau_0}\exp(-\frac{\pi}{4}\lambda + i\frac{\pi}{4} + iS(|\tau_0|)) \tag{2.73}$$

$$\phi_2(t_0) \sim \frac{1}{\gamma}\sqrt{2\beta}\exp\,(-\frac{\pi}{4}\lambda - i\frac{\pi}{4} - iS(|\tau_0|)), \tag{2.74}$$

and at $t \longrightarrow +\infty$:

$$\phi_1(t) \sim \frac{2}{|\gamma|}\sqrt{\beta\sinh\,(\pi\lambda)}\exp\,(\frac{\pi}{4}\lambda - i\frac{\pi}{2} - iS(\tau) - iarg\Gamma(i\lambda)) \tag{2.75}$$

$$\phi_2(t) \sim \frac{1}{\gamma}\sqrt{2\beta}\exp\,(\frac{3\pi}{4}\lambda - i\frac{\pi}{4} - iS(|\tau_0|)) \tag{2.76}$$

The self-consistent solution of the initial valued problem is as follows:

$$\begin{aligned}\phi_s(t\to\infty) &= \tfrac{|\gamma|}{\gamma}\sqrt{\exp(2\pi\lambda)-1}e^{\frac{-i3\pi}{4}-iS(|\tau_0|)-iS(\tau)-i\arg\Gamma(i\lambda)},\\ \phi_c(t\to\infty) &= e^{\pi\lambda+iS(|\tau_0|)-iS(\tau)}\end{aligned} \tag{2.77}$$

where $\tau = \sqrt{2\beta}t$, $S(\tau) = \tau^2/4 - \lambda\ln\tau$ and $\lambda = |\gamma|^2/(2\beta)$. Using this, one can derive

$$\begin{aligned}|\phi_s(t\to\infty)|^2 &= e^{2\pi\lambda} - 1\\ |\phi_c(t\to\infty)|^2 &= n_{sp} = e^{2\pi\lambda}\end{aligned} \tag{2.78}$$

If initially one has coherent atomic states $|\alpha\rangle|\beta\rangle$ where $\hat{a}|\alpha\rangle = \alpha|\alpha\rangle$ and $\hat{b}|\beta\rangle = \beta|\beta\rangle$ then

$$\begin{aligned}n_{st} &= |\alpha|^2e^{2\pi\lambda} + |\beta|^2(e^{2\pi\lambda}-1) + 2\sqrt{e^{2\pi\lambda}-1}e^{\pi\lambda}|\alpha||\beta|\times\\ &\quad\cos\left(\frac{3\pi}{4} + 2\;S(|\tau_0|) + \arg(\Gamma(i\lambda)) + \arg(\gamma) + \;arg(\alpha) + \arg(\beta)\right)\end{aligned} \tag{2.79}$$

so, depending on the initial arguments of α and β the number of produced particles due to stimulated transitions can be in the range $n_- < n_{st} < n_+$ where

$$n_\pm = \left(|\alpha|e^{\pi\lambda} \pm |\beta|\sqrt{e^{2\pi\lambda}-1}\right)^2 \tag{2.80}$$

Now, lets proceed to the problem of correlations between the two modes when initially there are no particles in atomic condensates. For this purpose consider the quadrature phase amplitudes:

$$\hat{X}_\theta(t) = (\hat{a}(t) + \hat{b}(t))e^{i\theta} + (\hat{a}^\dagger(t) + \hat{b}^\dagger(t))e^{-i\theta} \tag{2.81}$$

If initially the atomic condensates are in vacuum states then:

$$< \hat{X}_\theta{}^2(t) >= |\phi_c(t)e^{i\theta} + \phi_s^*(t)e^{-i\theta}|^2 \tag{2.82}$$

and long time after curve crossing event we obtain:

$$< \hat{X}_\theta{}^2(t \to \infty) >= |\sqrt{e^{2\pi\lambda}-1} + e^{\pi\lambda - \frac{i3\pi}{4} - i\arg\gamma - 2iS(\tau) - i\arg\Gamma(i\lambda) + 2\imath\theta}|^2 \tag{2.83}$$

Choosing two orthogonal phase angles $\theta_+ = 1/2(\frac{3\pi}{4} + \arg\gamma + 2S(\tau) + \arg\Gamma(i\lambda))$ and $\theta_- = 1/2(\frac{-\pi}{4} + \arg\gamma + 2S(\tau) + \arg\Gamma(i\lambda))$ we find:

$$< X^2_{\theta_\pm} >= |\sqrt{e^{2\pi\lambda}-1} \pm e^{\pi\lambda}|^2 \tag{2.84}$$

The result (2.84) means that dissociation forms atoms in a two mode squeezed state. Such correlated states have been under a strong interest in atomic physics [79], [86], [87].

If components A and B are distinct by some internal degree of freedom that is not conserved during the reaction then the Hamiltonian (2.56) should be generalized

so that reaction go in all possible channels:

$$\hat{H} = \mu_1(t)\hat{a}^\dagger\hat{a} + \mu_2(t)\hat{b}^\dagger\hat{b} + \gamma\hat{a}\hat{a} + \gamma^*\hat{a}^\dagger\hat{b}^\dagger + \gamma_a\hat{a}\hat{a} + \gamma_a^* a^\dagger a^\dagger + \gamma_b bb + \gamma_b^* b^\dagger b^\dagger \tag{2.85}$$

In this case even if one treats the field of initial condensate as a c-number, the resulting Landau-Zener problem in not analytically solvable, since all operator equations are no longer decoupled. However, if an additional symmetry exists, exact asymptotics can be found. For example, if the chemical potentials are equal due to some symmetry

$$\mu_1(t) = \mu_2(t) \tag{2.86}$$

then the equivalent Landau-Zener problem can be solved. For simplicity lets also assume that $\gamma_b = \gamma_a = \gamma_1$. Then the operator evolution is governed by the following equations:

$$\begin{aligned} i\dot{\hat{a}} &= t\hat{a} + \gamma b^\dagger - 2\gamma_1^*\hat{a}^\dagger \\ i\dot{\hat{b}} &= t\hat{b} + \gamma^* a^\dagger - 2\gamma_1^*\hat{b}^\dagger \end{aligned} \tag{2.87}$$

this is accomplished by the hermitian conjugate equations. By adding the equations in (2.87) and denoting $\hat{c} = \hat{a} + \hat{b}$ one gets

$$i\dot{\hat{c}} = t\hat{c} + (\gamma^* - 2\gamma_1^*)\hat{c}^\dagger \tag{2.88}$$

so that for $\hat{c}$ and $\hat{c}^\dagger$ the problem is reduced to already solved one. Accordingly, there are similar equations for operators $\hat{d}$ and $\hat{d}^\dagger$, where $\hat{d} = \hat{a} - \hat{b}$:

$$i\dot{\hat{d}} = t\hat{d} - (\gamma^* + 2\gamma_1^*)\hat{d}^\dagger \tag{2.89}$$

However, such a solution would be very unstable in respect of the terms that break the symmetry (2.86) since there are states that have the same energy participating in the curve crossing.

G. Landau-Zener transitions in a linear chain

In all models described above the number of independent states is finite but the limits of infinite number of states can be considered. In addition to those solvable classes there are models whose solution could be found so far only in the limit of infinite Hylbert space.

One such a class describes the dynamics of a system at an intersection of the two infinite bands of parallel levels with equal transition probabilities between any pair of states from different bands [36]. The physical application of this solution is obscure and the notion of transition probability matrix does not make sense there since transition probabilities do not saturate when time goes to infinity. Another class of multistate models with infinite number of states having well defined transition probabilities and clear physical interpretation was discovered by Porkrovsky and Sinitsyn in [41]. Physically its solution describes the quantum electron transfer between donor and acceptor separated by a long polymer strand (molecular bridge). The bridge can be considered as a linear array of identical sites. Such one-dimensional atomic-scale wires were intensely studied, both experimentally and theoretically [91, 92, 93]. Similar dynamics can be found in semiconductor superlattices [94, 95].

The goal is to describe the tunneling of a particle in such systems driven by a time-dependent homogeneous external field. An important assumption is that all molecular fragments in the chain are identical. Electric field splits the energy levels at different sites of the chain and suppresses the transitions. Hence electron hoppings proceed in a narrow time intervals close to moments at which electric field becomes zero. Since the tunneling is a fast process, one can disregard the relaxation originating from phonons and other elementary excitations.

Lets denote $|n\rangle$ a state located at the n-th site of the chain. These states form

a complete orthonormal set (Wannier basis). In terms of this set the electron Hamiltonian reads:

$$\hat{H} = \sum_{n=1}^{N} (\gamma \mid n\rangle\langle n+1 \mid + c.c.) + nF(t) \mid n\rangle\langle n \mid; \qquad (2.90)$$
$$F(t) = eE(t)a$$

where $E(t)$ is the electric field, e-electron charge, a is the distance between sites and γ is the coupling constant. A series of exact solutions of the time-dependent Shrödinger equation with the Hamiltonian (2.90) at $N = \infty$ known as drifting plane waves have been found long ago [95, 96, 97, 98, 99].

The set $|n\rangle$ is conventionally called the diabatic basis. The diabatic states are not eigenstates of the total Hamiltonian (2.90) with t or $E(t)$ considered as a parameter, they are eigenstates of the diagonal part of this Hamiltonian. Until $\mid F(t) \mid \gg \gamma$ the transitions between states are suppressed. This is the region of adiabatic regime. The adiabaticity is violated in a vicinity of the electric field nodes determined by inequality $\mid F(t) \mid \leq \gamma$, where all transitions proceed. The level crossing means that the diabatic levels cross, i.e. the diagonal elements of (2.90) coincide at $F(t) = 0$. The exact eigenvalues of the Hamiltonian (2.90) never cross according to the Wigner - Neumann theorem on avoided crossing. Suppose for definiteness that the node position in time is at $t = 0$. Since transitions are substantial only in the vicinity of the node, the exact dependence of the field on time can be reasonably approximated by linear one: $F(t) \approx \dot{F}(0) \cdot t$. At zero electric field E and free boundary conditions the Hamiltonian (2.90) can be easily diagonalized. Its spectrum is:

$$\varepsilon_j = 2g \cos(\pi j/N); \quad j = 1 \ldots N \qquad (2.91)$$

The matrix representation for the Hamiltonian (2.90) reads:

$$H_{nm} = n\dot{F}(0)t\delta_{nm} + \gamma(\delta_{m,n+1} + \delta_{m,n-1}) \tag{2.92}$$

For an infinite chain ($N \to \infty$) these equations are valid at any n and m. After a proper rescaling of time the Hamiltonian (2.92) becomes dimensionless:

$$H_{mn} = nt\delta_{mn} + g(\delta_{m,n+1} + \delta_{m,n-1}) \tag{2.93}$$

It depends on only one dimensionless number $g = \gamma/\sqrt{\dot{F}(0)}$, which is the Landau-Zener parameter. Corresponding system of Schrödinger equations for amplitudes $c_n(t)$ is:

$$i\dot{c}_n = ntc_n + g(c_{n-1} + c_{n+1}) \tag{2.94}$$

The problem is to find the transition matrix for this system, i.e. to find an asymptotic at $t \to +\infty$ of a solution $c_n(t)$ which obeys the initial condition $|c_n(t)|^2 = \delta_{n,n'}$ at $t \to -\infty$.

Lets introduce an auxiliary function $u(\varphi, t) = \sum_{n=-\infty}^{\infty} c_n(t)e^{in\varphi}$. The system (2.94) is equivalent to a following equation in partial derivatives for $u(\varphi, t)$:

$$\frac{\partial u}{\partial t} + t\frac{\partial u}{\partial \varphi} + 2igu\cos\varphi \;=\; 0 \tag{2.95}$$

One should find a solution of this equation which obeys the initial condition: $u(\varphi, t) \to \exp[in'(-\frac{t^2}{2} + \varphi)]$ at $t \to -\infty$. Given the solution $u(\varphi, t)$, the amplitudes $c_n(t)$ can be found by the inverse Fourier transformation: $c_n(t) = \frac{1}{2\pi}\int_0^{2\pi} u(\varphi, t)e^{-in\varphi}d\varphi$. The solution of eq. (2.95) which obeys proper boundary conditions is:

$$u(\varphi, t) = \exp\left[-i\left(2g\int_{-\infty}^{t}\cos\left(\varphi - \frac{t^2}{2} + \frac{t'^2}{2}\right)dt' + n'\left(\varphi - \frac{t^2}{2}\right)\right)\right] \tag{2.96}$$

Putting $t = +\infty$ in the solution (2.96) and taking the inverse Fourier-transform, one can arrive at following asymptotics:

$$c_n(t) \approx \exp\left(-\frac{int^2}{2} + i\frac{(n'-n)\pi}{4}\right) J_{|n-n'|}(2\sqrt{2\pi}g) \tag{2.97}$$

Thus, the scattering amplitudes in terms of modified states, with the fast phase factor $\exp(-int^2/2)$ incorporated, are:

$$\langle n \mid U_\infty \mid n' \rangle = e^{i(n'-n)\pi/4} J_{|n-n'|}(2\sqrt{2\pi}g) \tag{2.98}$$

here U_∞ is an evolution operator from $t \to -\infty$ to $t \to +\infty$ It displays infinite number of oscillations with the LZ parameter g. However, for large $|n-n'|$ the oscillations start with $g > |n-n'|$. These oscillations can be observed experimentally by varying the field sweep rate $\dot{E}(0)$. For small values of g the amplitudes are small and quickly decreasing with $|n-n'|$ growing.

For large $g \gg |n-n'|$ the asymptotics of the amplitudes (2.98) are:

$$\langle n \mid U \mid n' \rangle \sim \frac{e^{i(n'-n)\pi/4}}{(\sqrt{2\pi^3}g)^{1/2}} \cos(2\sqrt{2\pi}g - \frac{(n-n')\pi}{2} - \frac{\pi}{4}) \tag{2.99}$$

The asymptotics in the model with infinite number of states at large coupling constant can be very different from those in models with finite number of states. For finite Hylbert space in the limit $g \gg N$, where N is the number of intersecting states, transition probabilities behave as exponents $\exp(-C(n,n')g^2)$ where $C(n,n')$ do not depend on g. In contrast the result (2.99) displays power law with oscillations instead of exponential dependence of transition probabilities on g for large g. This is the result of quantum interference of different Feynman trajectories. A step in this trajectory has average length g (see below). Such a step can not be realized in a system with a finite number of states when $g \gg N$.

The mean square of displacement at one crossing event is:

$$< (n-n')^2 >= \sum_{n=-\infty}^{\infty} (n-n')^2 |J_{|n-n'|}(2\sqrt{2\pi}g)|^2 = \\ = 4\pi g^2 \tag{2.100}$$

If the external field is periodic in time and the coherence between crossing events is lost, the electron performs a random walk, i.e. it diffuses. Suppose the field changes harmonically $F(t) = F_0 \sin(\omega t)$. At time points $t_k = \pi k/\omega$ (k is an integer) all levels cross at one point. The square of Landau-Zener parameter is $g^2 = \frac{\gamma^2}{F_0 \omega}$. The diffusion coefficient is $D = 2a^2 < (n-n')^2 > /T$, where $T = 2\pi/\omega$ is the period of oscillations and factor 2 accounts for two crossing events per period. Collecting these results and equation (2.100), we find:

$$D = \frac{4a\gamma^2}{F_0} \tag{2.101}$$

This result does not depend on the frequency of the external field.

CHAPTER III

NOISE IN THE LANDAU-ZENER TRANSITIONS

Emerging new applications of the Landau-Zener theory raise the question of the applicability of the Landau-Zener formula when the coupling of quantum states to environment becomes unavoidable. The effect of environment on the Landau-Zener transition probability can be studied by additional coupling of the two level system to the phonon or to the spin bath. However the resulting Hamiltonians become very complicated and usually do not allow analytical predictions in wide range of parameters. One of the possible approaches to the coupling to a bath problem is to reformulate it in terms of the multistate Landau-Zener model [61, 56]. This approach was proved to be useful in application to nanomagnets coupled to the nuclear spin bath and to coupling to phonon bath at low temperature. It can be justified mainly when the internal dynamics of the bath is not important in comparison with dynamics due to its coupling to the central two level system. In the opposite limit the bath has strong its own dynamics which is mainly not influenced by the coupling to the central system. The action of such a bath on the two-state system can be modeled as the action of a stationary and usually fast noisy field [49]. If this noise is generated by many sources it can be considered as Gaussian.

Landau-Zener transitions in a two-level system with a non-diagonal noise were studied for the first time by Kayanuma in [47]. The Hamiltonian of the problem was chosen to be

$$\hat{H} = \beta t\hat{\sigma}_z + \eta_x\hat{\sigma}_x \tag{3.1}$$

where η_x is the noise field with the correlation function $\langle\eta_x(t_1)\eta_x(t_2)\rangle = J_x^2 e^{-\lambda_x|t_1-t_2|}$ and $\hat{\sigma}_i$ are Pauli matrices. Note that the constant transverse field term was not considered by Kayanuma. In the limit of infinitely short correlation time $\lambda \to \infty$ he found

a simple analytical result for the transition probability. The approach of Kayanuma was to write the probability as a series in powers of noisy field. For exponentially correlated noise all integrals in such a series are Gaussian; this strongly simplified the analysis of the series but restricted the result to exponentially correlated noise only. Pokrovsky and Sinitsyn [100, 101] generalized the Kayanuma model introducing a more general Hamiltonian with all three components of random magnetic field being non-zero and possibly not equal and with the most general form of the short-time correlation tensor. The regular constant transverse magnetic field was also included. Moreover, the Hamiltonian of the Landau-Zener transitions in the two level system is equivalent to the one for a spin-1/2 evolution in the time-dependent magnetic field. The generalization of this problem to higher spins is one of the most useful multistate Landau-Zener models therefore most of the calculations in this section will be done for the case of an arbitrary spin ($S \geq 1/2$) with the Hamiltonian

$$\hat{H} = (\beta t)\hat{S}_z + g\hat{S}_x + \sum_{i=-1}^{+1} \eta_i \hat{S}_i, \;\; i = x, y, z \tag{3.2}$$

$$\langle \eta_i(t_1)\eta_j(t_2)\rangle = f_{ij}(\lambda|t_1 - t_2|) \tag{3.3}$$

The second term in the Hamiltonian (3.2) can be considered as a constant transverse magnetic field acting on a spin. The measurements of the LZ transition probabilities can provide an information about the strength of the coupling to the bath. One of such examples is a molecular nanomagnet in fluctuating dipole field. The Hamiltonian (3.2) may be relevant to the quantum shuttle problem where avoided level crossings occurred to be important [14]. The fast noise in this example corresponds to thermal fluctuations. Adiabatic avoided crossings were proposed recently for generation of entangled states of q-bits [24]. Then the understanding of the noise

effects is important to trace the error propagation in a quantum circuit.

The result of the evolution with random field depends on a given noise realization. Hence instead of looking for a solution of the Schrödinger equation with a special time-dependence of the noisy field, one should find the transition probabilities averaged over noise realizations, namely

$$P_{j\leftarrow i} = \langle |a_j(t \to \infty)|^2 \rangle_{noise} \tag{3.4}$$

where $a_k(t)$ is the amplitude of the k-th eigenstate of $\hat{S}_z$-operator and $|a_i(t \to -\infty)|^2 = 1$

A. Density matrix and Bloch tensors

The Hamiltonian (3.2) contains only operators that belong to the SU(2) algebra. In terms of components with definite projections it reads (note that $b_\pm = b_x \pm i b_y$; $S_\pm = S_x \pm i S_y$):

$$H = -b_z S_z - \frac{1}{2}(b_+ S_- + b_- S_+) \tag{3.5}$$

where b_z and $b_\pm$ are some time-dependent coefficients.

When the random magnetic field acts onto the spin, it must be described by the density matrix $\widehat{\rho}$. By definition it is a $(2S+1) \times (2S+1)$ Hermitian matrix with the trace equal to 1. It satisfies the standard equation of motion:

$$i\frac{d\widehat{\rho}}{dt} = [H, \widehat{\rho}] \tag{3.6}$$

Any Hermitian matrix with the trace equal to 1 can be represented as a sum:

$$\widehat{\rho} \;=\; \frac{1}{2S+1}\hat{I} + \mathbf{g}\cdot\hat{\mathbf{S}} + \frac{1}{2}g_{ik}\left(\hat{S}_i\hat{S}_k + \hat{S}_k\hat{S}_i - \frac{2}{3}\delta_{ik}S(S+1)\right) + \tag{3.7}$$

$$...\frac{1}{(2S)!}g_{i_1 i_2 ... i_{2S}}\left(\hat{S}_{i_1}\hat{S}_{i_2}...\hat{S}_{i_{2S}} + \text{all permutations} - \text{all traces}\right) \qquad (3.8)$$

If the Hamiltonian is the generator of rotation (which is true in our case), each term in equation (3.8) corresponds to an irreducible representation and evolves independently. The symmetric tensors g_{ik}, g_{ikl} $...g_{i_1 i_2 ... i_{2S}}$ are called the Bloch tensors in analogy with the Bloch vector $\mathbf{g}$ well known from Bloch theory of the nuclear spin motion. Any trace of such a tensor must be equal to zero. The Hamiltonian (3.2) generates following equations of motion for the Bloch tensors:

$$\dot{\mathbf{g}} = -\mathbf{b} \times \mathbf{g}; \quad \dot{g}_{ik} = -\varepsilon_{ilm} b_l g_{mk} - \varepsilon_{klm} b_l g_{im}; ... \qquad (3.9)$$

All these equations are independent and have obvious integrals of motion:

$$\mathbf{g}^2 = const; \; g_{ik}^2 = const; \; g_{ikl}^2 = const; ... \qquad (3.10)$$

Thus, the density matrix of a spin S in an external time-dependent magnetic field has $2S$ conserving values. It is convenient to represent the Bloch tensors by their complex components with a definite projection to the $z-$axis. We will denote such components of a tensor of the rank s as $g_{s,m}$. The corresponding tensor operators composed from the symmetrized products of $2s$ components of the spin S operators are denoted $\hat{T}^S_{s,m}$. They can be constructed from the senior operator of this representation $\hat{T}^S_{s,s} = 2^{-s/2}\hat{S}^s_+$ with $\hat{S}_\pm = \hat{S}_x \pm i\hat{S}_y$ by recurrent commutations with the operator S_-:

$$\hat{T}^S_{s,m} = -\frac{1}{\sqrt{(s+m+1)(s-m)}}\left[\hat{S}_-, \hat{T}^S_{s,m+1}\right] \qquad (3.11)$$

The operators $\hat{T}^S_{s,m}$ are polynomials of the standard spin operators $\hat{S}_\pm$ and $\hat{S}_z$. They are operator analogs of spherical harmonics. We show below relations between the Cartesian components of the tensor $g_{i_1 ... i_s}$ and its components $g_{s,m}$ for several values

of s:

$$g_{1,\pm1} = \frac{1}{\sqrt{2}}\left(g_x \pm i g_y\right);\ g_{1,0} = g_z \tag{3.12}$$

$$g_{2,\pm2} = \frac{1}{\sqrt{6}}\left(g_{xx} - g_{yy} \pm 2i g_{xy}\right);\ g_{2,\pm1} = \frac{1}{\sqrt{2}}\left(g_{xz} \pm i g_{yz}\right); g_{2,0} = g_{zz} \tag{3.13}$$

$$g_{3,\pm3} = \frac{1}{\sqrt{20}}\left(g_{xxx} \pm 3i g_{xxy} - 3g_{xyy} \mp i g_{yyy}\right); ... \tag{3.14}$$

The general rule for writing the $s, \pm m-$component via its Cartesian counterparts is the same as for the product $\frac{m!}{\sqrt{2m!}}(x \pm iy)^m z^{s-m}$. Equations (3.9) in terms of the components with definite $z-$projections become:

$$\dot{g}_{s,m} = -imb_z g_{s,m} + \frac{i}{2}\sqrt{(s+m)(s-m+1)}b_+ g_{s,m-1} + \frac{i}{2}\sqrt{(s-m)(s+m+1)}b_- g_{s,m+1} \tag{3.15}$$

and the conservation laws are:

$$\sum_{m=-s}^{s} |g_{s,m}|^2 = const \tag{3.16}$$

B. Fast noise in a two-level system

Consider only a spin 1/2 or, equivalently a two-level system. Assume that the magnetic field can be separated into regular and random parts:

$$\mathbf{b}(t) = \mathbf{b}_r(t) + \eta(t) \tag{3.17}$$

where $\mathbf{b}_r(t) = \hat{z}\dot{b_z}t + \hat{x}b_x$ and $\eta(t)$ is the Gaussian noise determined by its correlators:

$$\langle \eta_i(t)\eta_k(t')\rangle = f_{ik}(t-t') \tag{3.18}$$

Assume also that the correlators (3.18) decay after a characteristic time difference τ_n and that this correlation time is much less than the characteristic time of the LZ process τ_{LZ}. However, the noise must be slow enough to avoid the direct transitions between the levels when the interlevel distance approaches its saturation or characteristic value ω far from the crossing point. Thus, the noise correlation time τ_n must satisfy a following inequalities:

$$\omega^{-1} \ll \tau_n \ll \tau_{LZ} \tag{3.19}$$

The spectral width of noise is $1/\tau_n$. The noise produces transitions during the interval of time $t_{acc} = 1/(\dot{b}_z \tau_n)$, after which the current LZ frequency becomes larger than the noise spectral width. This interval is called the accumulation time. It is much larger than other characteristic time intervals τ_n and τ_{LZ}.

Consider first an auxiliary problem in which $b_x = 0$ and transitions are mediated by noise only. Such a problem for a special shape of correlators ($f_{xx} = J^2 \exp\left(-\frac{t-t'}{\tau_n}\right)$; the remaining components of the correlation tensor are zero) was solved earlier by Kayanuma [45, 47] and studied numerically by Nishino *et al.* [48]. Pokrovsky and Sinitsyn [100] have generalized the Kayanuma solution. Equations for the components of the Bloch vector in this case are:

$$\dot{g}_z = (i/\sqrt{2})\left(\eta_+ g_- - \eta_- g_+\right); \;\; \dot{g}_\pm = \mp i\left(\dot{b}_z t + \eta_z\right) g_\pm + (i/\sqrt{2})\eta_\pm g_z \tag{3.20}$$

Solving equation for $g_\pm$, we find:

$$g_\pm(t) = g_\pm(-\infty)\exp\left(\mp\frac{i\dot{b}_z t^2}{2} \mp i\int\limits_{-\infty}^{t}\eta_z(t')dt'\right) + \tag{3.21}$$

$$(i/\sqrt{2})\int\limits_{-\infty}^{t}\exp\left[\mp\frac{i\dot{b}_z(t^2-t'^2)}{2} \mp i\int\limits_{t'}^{t}\eta_z(t'')dt''\right]\eta_\pm(t')g_z(t')dt' \tag{3.22}$$

Let us first consider the case of complete initial decoherence: $g_{\pm}(-\infty) = 0$. Then, plugging equation (3.22) into the first equation (3.20), we find a separate equation for g_z:

$$\dot{g}_z = -(1/2) \int_{-\infty}^{t} \exp\left[-\frac{i\dot{b}_z(t^2 - t'^2)}{2} - i\int_{t'}^{t} \eta_z(t'')dt''\right] \eta_+(t)\eta_-(t')g_z(t')dt' + c.c. \quad (3.23)$$

Let us average equation (3.23) over the ensemble of the random noise. For such averaging it is important that the noise correlation time τ_n is much shorter than the time t_{acc} necessary for a substantial variation of $\langle g_z \rangle$. This fact allows to represent the average $\langle \eta_+(t)\eta_-(t')g_z(t') \rangle$ approximately as a product: $\langle \eta_+(t)\eta_-(t')g_z(t') \rangle \approx \langle \eta_+(t)\eta_-(t') \rangle \langle g_z(t') \rangle$. More accurately one should incorporate the fluctuations of g_z. In the leading approximation they are determined by the same equation (3.23) as follows:

$$\delta g_z = -(1/2) \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} e^{-\frac{i\dot{b}_z(t_1^2 - t_2^2)}{2}} \left(\eta_+(t_1)\eta_-(t_2) - \langle \eta_+(t_1)\eta_-(t_2) \rangle\right) \langle g_z(t_2) \rangle \, dt_2 + c.c \quad (3.24)$$

We ignore η_z (this approximation will be justified by the next step). Let substitute this additional term into equation (3.23) and perform averaging over the gaussian random field η. According to the Wick's rule, it is reduced to all possible pairings. In our case the only possible pairing is $\langle \eta_+(t)\eta_-(t_2) \rangle \langle \eta_-(t')\eta_+(t_1) \rangle$. Such a pairing limits the integration by the interval $t - \tau_n < t_2 < t_1 < t' < t$. Thus the contribution of the fluctuational term differs by an additional factor $\sim \tau_n / t_{acc} << 1$ from the principal contribution from $\langle g_z \rangle$. These arguments represent a shortened version of the original arguments by Kayanuma [45] and are akin to the Abriikosov-Gor'kov theory of static disordered alloys [88].

Using the fact that the decay of the correlator $\langle \eta_+(t)\eta_-(t') \rangle$ limits effectively the integration over time by an interval $t - \tau_n < t' < t$, we can prove that the contribution

of the noise component η_z in the exponent in the equation (3.23) can be neglected. To estimate this contribution we assume that η_z is statistically independent from other components. Then the averaging over η_z results in the Debye-Waller factor $\exp\left[-\frac{1}{2}\left\langle\left(\int_{t'}^{t}\eta_z(t'')dt''\right)^2\right\rangle\right]$. The argument of this exponent can be estimated as $\langle\eta_z^2\rangle\,\tau_n^2$. It is small provided the level of noise $\langle\eta_z^2\rangle$ is much smaller than τ_n^{-2}. The noise correlators are even functions of the time difference. Therefore, expanding linearly the time argument of the exponent in the same equation $\frac{\dot{b}_z(t^2-t'^2)}{2}\approx\dot{b}_z t(t-t')$, one can transform the integral-differential equation (3.23) into an ordinary differential equation for $\langle g_z\rangle$:

$$\dot{\langle g_z\rangle} = -\hat{F}\left(\dot{b}_z t\right)\langle g_z\rangle \tag{3.25}$$

where $\hat{F}(\Omega)$ is the Fourier-transform of the function $F(\tau)=f_{xx}(\tau)+f_{yy}(\tau)$:

$$\hat{F}(\Omega)=\int_{-\infty}^{\infty}F(\tau)\cos\Omega\tau d\tau \tag{3.26}$$

Equation (3.25) has a simple solution:

$$\langle g_z(t)\rangle=\langle g_z(-\infty)\rangle\exp\left[-\int_{-\infty}^{t}\hat{F}\left(\dot{b}_z t'\right)dt'\right] \tag{3.27}$$

At $t\to+\infty$ the asymptotic value of $\langle g_z\rangle$ is:

$$\langle g_z(+\infty)\rangle=\langle g_z(-\infty)\rangle\exp\left(-\theta\right);\quad \theta=\frac{\pi F(0)}{\dot{b}_z} \tag{3.28}$$

Note that what matters for the LZ transition is the average quadratic fluctuation of non-diagonal noise at any moment $F(0)=\left\langle\eta_x^2+\eta_y^2\right\rangle$ in contrast to a standard characteristic of the white noise which would be $\zeta=\hat{F}(0)/(2\lambda)$. Indeed, commonly white noise correlator is introduced as $\langle\eta(t)\eta(t')\rangle=\gamma\delta(t-t')$. The only characteristic of the noise in this approach is $\gamma=\int_{-\infty}^{\infty}\langle\eta(t)\eta(t')\rangle\,dt'$. An interesting feature of the asymptotic formula (3.28) is its independence on the noise correlation time τ_n.

However, it should be kept in mind that this asymptotic is valid only at time $t \gg t_{acc} = \left(\dot{b}_z \tau_n\right)^{-1}$.

The term $g_{\pm}(-\infty) \exp\left(\mp\frac{i\dot{b}_z t^2}{2} \mp i \int\limits_{-\infty}^{t} \eta_z(t')dt'\right)$, omitted at substitution in the first equation (3.20) and the ensemble averaging, vanishes if the $z-$component of the noise is statistically independent from others. Now let us perform a similar procedure solving first equation for g_z and then substituting the solution into equations for $g_{\pm}$. In the same approximation equation for averages of these components of the Bloch vector reads:

$$\langle \dot{g}_{\pm} \rangle = -\frac{1}{2}\hat{F}\left(\dot{b}_z t\right) \langle g_{\pm} \rangle \tag{3.29}$$

The first term in equation (3.22) after the averaging turns into zero at any finite t. Indeed the Debye-Waller factor which appears in this case is $\exp\left[-\frac{1}{2}\left\langle\left(\int\limits_{t}^{\infty} \eta_z(t'')dt''\right)^2\right\rangle\right] = 0$. The asymptotics of $\langle g_{\pm} \rangle$ at $t \to +\infty$ are:

$$\langle g_{\pm}(+\infty) \rangle = \exp\left(-\frac{\theta}{2}\right) \langle g_{\pm}(-\infty) \rangle\,; \quad \theta = \frac{\pi F(0)}{\dot{b}_z} \tag{3.30}$$

Note that the symbols $g_{\pm}(\pm\infty)$ denote the coefficients at $\exp\left(\mp\frac{i\dot{b}_z t^2}{2}\right)$. Thus, the noise asymptotically tends to reduce the average components of the Bloch vector, i.e. to establish equipopulation of the levels and to destroy the coherence. However, during the time interval of the order of $t_{acc}$the average Bloch vector can oscillate.

For the considered problem it is possible to find exactly the fluctuations of the Bloch vector. Indeed, they are given by a standard formula:

$$\left\langle (\delta \mathbf{g})^2 \right\rangle = \left\langle \mathbf{g}^2 \right\rangle - \langle \mathbf{g} \rangle^2 \tag{3.31}$$

Due to the symmetry of the problem $\mathbf{g}^2$ is a conserving value. Therefore, its average coincides with itself and is determined by the initial conditions. The average value

of the vector $\langle \mathbf{g} \rangle$ was calculated above. Thus, we find an asymptotic value of the fluctuations:

$$\left\langle (\delta \mathbf{g})^2 \right\rangle \Big|_{t=+\infty} = g_z^2(-\infty)\left(1-\exp\left(-2\theta\right)\right) + \left[g_x^2(-\infty)+g_y^2(-\infty)\right]\left(1-\exp\left(-\theta\right)\right) \tag{3.32}$$

The values of average square fluctuation can be also written for any moment of time. The fluctuations are strong, i.e. their magnitude is of the same order as the average values of the Bloch vector components unless θ is very small. An important property of the noise is that in fluctuations it mixes diagonal and non-diagonal elements of the density matrix, i.e all three components of the Bloch vector. Correlations in the noisy LZ theory were studied in more details in [102].

C. Fast noise at a multilevel crossing

We consider only the case of a Zeeman multiplet placed into a varying magnetic field. It is described by equations (3.15). In this section we neglect the transitions produced by the regular part of the magnetic field and take in account only the transitions produced by the random field. This approach is correct outside the time interval τ_{LZ} near the avoided crossing point $t = 0$.

Suppose that initially all components of the s-tensor except of $g^S_{s,m}$ are zero. Then the chain of equations (3.15) can be truncated leaving only equations for $g_{s,m}$ itself and its nearest neighbors $g_{s,m\pm1}$. Others become vanishing in the fast noise limit. The truncated equations read:

$$\begin{aligned}
\dot{g}^S_{s,m} &= -im\dot{b}_z t g^S_{s,m} + i\left(\lambda_{s,m}\eta_+ g^S_{s,m-1} + \lambda_{s,-m}\eta_- g^S_{s,m+1}\right); \\
\dot{g}^S_{s,m-1} &= -i\left(m-1\right)\dot{b}_z t g^S_{s,m-1} + i\lambda_{s,m}\eta_- g^S_{s,m} \\
\dot{g}^S_{s,m+1} &= -i\left(m+1\right)\dot{b}_z t g^S_{s,m+1} + i\lambda_{s,-m}\eta_+ g^S_{s,m}
\end{aligned} \tag{3.33}$$

To deal with slow-varying average values the fast oscillating exponent should be eliminated. In order to do that we introduce the slow variable $\tilde{g}^S_{s,m} = g^S_{s,m} \exp\left(\frac{i\dot{b}_z t^2}{2}\right)$. After elimination of the values $g^S_{s,m\pm1}$ and averaging, we find a following equation for $g^S_{s,m}$:

$$\left\langle \dot{g}^S_{s,m} \right\rangle = -\frac{1}{2}\left[\left(s(s+1) - m^2\right)\hat{F}(\dot{b}_z t) + m\hat{G}(\dot{b}_z t)\right]\left\langle g^S_{s,m} \right\rangle \tag{3.34}$$

where $\hat{F}(\Omega)$ is defined by equation (3.26) and $\hat{G}(\Omega)$ is defined as a sine Fourier-transform:

$$\hat{G}(\Omega) = \int\limits_{-\infty}^{\infty} \langle \eta_x(\tau)\eta_y(0) - \eta_y(\tau)\eta_x(0)\rangle \sin\Omega t \; dt \tag{3.35}$$

Thus, the time dependence of the average $\left\langle g^S_{s,m} \right\rangle$ is defined as:

$$\left\langle g^S_{s,m}(t) \right\rangle = \exp\left\{-\frac{1}{2}\int\limits_{-\infty}^{t}\left[\left(s(s+1) - m^2\right)\hat{F}(\dot{b}_z t') + m\hat{G}(\dot{b}_z t')\right]dt'\right\}\left\langle g^S_{s,m}(-\infty) \right\rangle \tag{3.36}$$

Its asymptotic value at $t \to +\infty$ does not contain the sine Fourier-transform:

$$\left\langle g^S_{s,m}(+\infty) \right\rangle = \exp\left[-\frac{1}{2}\left(s(s+1) - m^2\right)\theta\right]\left\langle g^S_{s,m}(-\infty) \right\rangle \tag{3.37}$$

At $s = 1, m = 1$ this result coincides with (3.29).

Assuming the complete initial decoherence, only diagonal density matrix elements, and hence $g_{s,0}$, can be nonzero. For their average we find following equations:

$$\left\langle g^S_{s,0}(+\infty) \right\rangle = \exp\left[-\frac{s\,(s+1)}{2}\theta\right]\left\langle g^S_{s,0}(-\infty) \right\rangle \tag{3.38}$$

Note that these relations do not contain S explicitly, the time evolution depends on s only.

In the following tables we demonstrate the results of calculations according to this algorithm for the values of S 1 and 3/2 (the results for $S = 1/2$ are given in the

previous section by equation (3.55)).

$$
\begin{array}{cccc}
{}_j\backslash^{j'} & +1 & 0 & -1 \\
+1 & \frac{1}{3}+\frac{1}{2}E_1+\frac{1}{6}E_2 & \frac{1}{3}-\frac{1}{3}E_2 & \frac{1}{3}-\frac{1}{2}E_1+\frac{1}{6}E_2 \\
0 & & \frac{1}{3}+\frac{2}{3}E_2 & \\
-1 & & &
\end{array}
\tag{3.39}
$$

$$
\begin{array}{ccccc}
{}_j\backslash^{j'} & 3/2 & 1/2 & -1/2 & -3/2 \\
3/2 & \frac{1}{4}+\frac{9E_1}{20}+\frac{E_2}{4}+\frac{E_3}{20} & \frac{1}{4}+\frac{3E_1}{20}-\frac{E_2}{4}-\frac{3E_3}{20} & \frac{1}{4}-\frac{3E_1}{20}-\frac{E_2}{4}+\frac{3E_3}{20} & \\
1/2 & & \frac{1}{4}+\frac{E_1}{20}+\frac{E_2}{20}+\frac{9E_3}{20} & \frac{1}{4}-\frac{E_1}{20}+\frac{E_2}{20}-\frac{9E_3}{20} & \\
-1/2 & & & & \\
-3/2 & & & &
\end{array}
\tag{3.40}
$$

Here we denoted $E_s = \exp\left(-\frac{s(s+1)}{2}\theta\right)$, i.e. $E_1 = e^{-\theta};\ E_2 = e^{-3\theta};\ E_3 = e^{-6\theta};\ E_4 = e^{-10\theta}$. Unfilled sites in the table can be easily restored using the time reversal symmetry: $P_{j\to j'} = P_{j'\to j} = P_{-j\to -j'}$. In Fig.8 I provide results of the numerical test of the theoretical predictions for a 3-level system ($S = 1$). Theoretical and numerical predictions are in excellent agreement with each other.

The quadratic fluctuations of the values $g^S_{s,m}$ for fixed S and s are calculated as it was done in the previous section:

$$
\left\langle \delta \sum_{m=-s}^{s} \left| g^S_{s,m} \right|^2 \right\rangle = \sum_{m=-s}^{s} \left| g^S_{s,m} \right|^2 \Bigg|_{t=-\infty} - \left\langle g^S_{s,m} \right\rangle^2 \tag{3.41}
$$

In the case of complete initial decoherence employing equation (3.56), we find:

$$
\left\langle \delta \sum_{m=-s}^{s} \left| g^S_{s,m} \right|^2 \right\rangle_{t=+\infty} = \left[g^S_{s,0}(-\infty) \right]^2 \left[1 - E_s^2 \left(P_s^{0,0} \left(2e^{-2\pi\gamma^2} - 1 \right) \right)^2 \right] \tag{3.42}
$$

D. Transition time for colored noise

Lets proceed to the solution of a more general problem retaining non-zero $x-$component of the regular field b_x. This solution is possible because of large time-scales difference between processes of noise accumulation and conventional Landau-Zener transitions.

The typical time for establishing the asymptotic of the transition probabilities is $\lambda/\beta F(0)$. One can analyze the behavior of transitions driven by fast noise approximating it by the standard δ-like white noise in order to find typical transition rates and times. If the action of the standard white noise is limited by some finite time interval, it becomes physically equivalent to the fast noise with a finite amplitude.

Consider isotropic white noise $\langle \eta_i(t)\eta_k(t')\rangle = \gamma\delta_{ik}\delta(t-t')$. The application of the averaging technique designed above leads to a considerably simpler effective master equation of motion for averaged over noise Bloch tensors, for example, after an additional unitary transformation, the Bloch vector components satisfy the following equations [100]:

$$\dot{\langle g_i\rangle}_{noise} = -\gamma\langle g_i\rangle_{noise} \tag{3.43}$$

with a simple solution

$$\langle g_i(t)\rangle_{noise} = \langle g_i(t=t_0)\rangle exp(-\gamma t) \tag{3.44}$$

The paradox is that at $t\to\infty$ the formula (3.44) always leads to the occupation numbers $p=1/2$ but as follows from the above sections this does not happen for the fast but colored noise with a finite amplitude. The reason is that in the LZ model the solution strongly oscillates with the frequency roughly $\omega(t)\sim\beta t$ long before and after the level crossing point. This introduces a new energy scale that must be compared with λ. For time in the range $|\beta t|<\lambda$, the approximation of white noise is roughly valid even for finite noise amplitude, but beyond this interval of time the oscillations

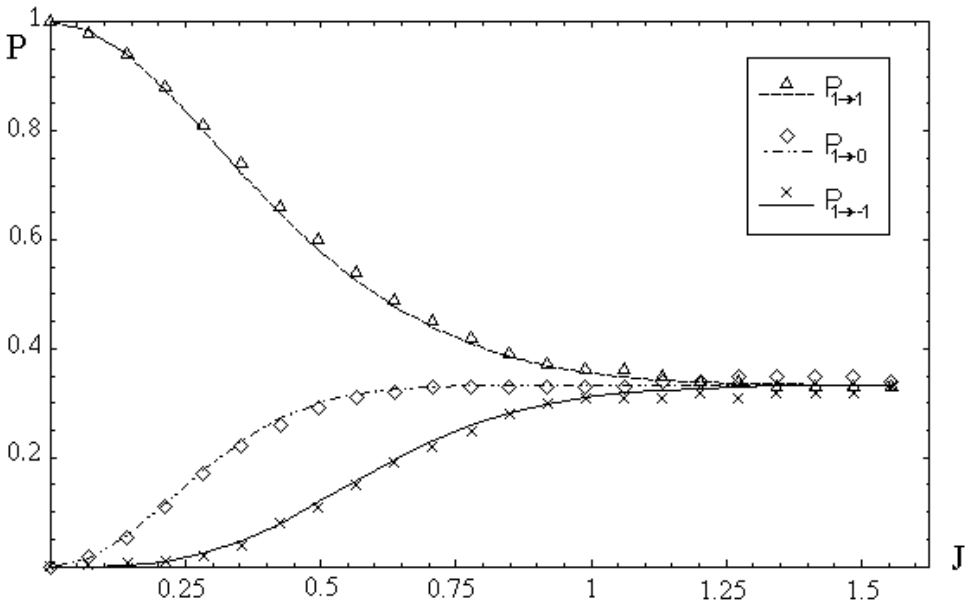

Fig. 8. The final probability to find the spin-1 system in the state with $S_z = -1$, 0 or 1 when the initial state is $S_z = 1$ as function of the noise amplitude. The Hamiltonian is $H = tS_z + \eta_x S_z$ where $\langle \eta_x(t_1)\eta(t_2)\rangle = Je^{-\lambda|t_1-t_2|}$. Discrete points correspond to results of numerical simulations with averaging over 200 different noise realizations and $\lambda = 125$. Lines correspond to analytical predictions of (3.39).

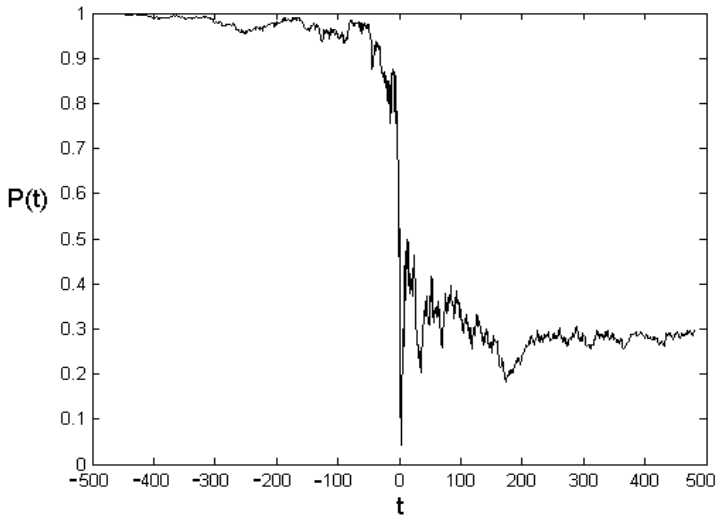

Fig. 9. Typical evolution of the probability $P(t) = |a_1(t)|^2$, where $a_1(t)$ is the amplitude of the first diabatic state. The choice of parameters is as follows, $S = 1/2$, $f_{ij} = \delta_{ij} J^2 e^{-\lambda|t-t'|}$, $\beta = 1$, $\lambda = 80$, $J^2 = 0.18$, $g = 0.7$.

of the LZ solution become faster than the correlation time of the noise, and the action of the noise becomes suppressed by the oscillations.

In Fig.9 a typical evolution of the probability is shown for a two level system to stay in the same state as a function of time. The evolution reminds diffusive motion that slowly stops at large absolute values of time. The sharp change of the transition probability near $t = 0$ is due to the action of the regular component of the transverse field.

To estimate roughly the transition probability due to the noise action one can apply the standard white noise approximation in the time interval $|t| < C\lambda/\beta$ and accept that at $|t| > C\lambda/\beta$ no transitions due to the noise happen. The parameter C is a constant of the order of unity. The result is $\rho(\infty) = \exp(-2C\lambda\gamma/\beta)\rho(-\infty)$. According to the definition, $\gamma \sim F(0)/(2\lambda)$ in agreement with the calculations of the previous sections. However the calculation of the coefficient C (and proof that

it is independent on parameters of the Hamiltonian) is important especially since it appears in the exponent. Summarizing, the transitions mediated by the fast noise proceed during the large time interval of the order of λ/β. This is the reason why its total effect remains finite, though it is very small on a typical time scale of the usual LZ-transitions mediated by constant field ($\tau \sim \Gamma/\beta$) [89, 90].

E. Landau-Zener transitions in a constant transverse field and in the colored noise

The solution of a more general problem retaining non-zero $x-$component of the regular field b_x becomes possible due to separation of times [58]: one can neglect the effect of b_x beyond the LZ time interval τ_{LZ} near the crossing point. On the other hand, one can neglect the effect of the noise inside this and even much larger interval of time, since its characteristic time is $t_{acc} \gg \tau_{LZ}$. Thus, the problem is separated into 3 parts: in two intervals $(-\infty, -t_0)$ and $(t_0, +\infty)$ we can use the solution of our auxiliary problem, i.e. to take in account only transitions caused by the noise; in the interval $(-t_0, t_0)$ the Landau-Zener solution is valid. One needs only to match them properly. The time interval t_0 must satisfy a strong inequality: $\tau_{LZ} \ll t_0 \ll t_{acc}$. As it has been shown before, the average components g_z and $g_\pm$ evolve separately under the action of the noise produced transitions. Therefore, at the moment of time $-t_0$ these components are:

$$\langle g_z(-t_0)\rangle = \exp(-\theta/2) g_z(-\infty); \quad \langle g_\pm(-t_0)\rangle = \exp(-\theta/4) g_\pm(-\infty) \tag{3.45}$$

The action of the LZ transition matrix (1.3) with the matrix elements (2.26) can be transferred onto the vector $\mathbf{g}$ by using spin-1 matrix (2.27). For simplicity we write the result for the component g_z:

$$\langle g_z(t_0)\rangle = \sqrt{2}ab^* \langle g_+(-t_0)\rangle + (2|a|^2-1)\langle g_z(-t_0)\rangle + \sqrt{2}a^* b \langle g_-(-t_0)\rangle \quad (3.46)$$

$$\langle g_+(t_0)\rangle = a^2 \langle g_+(-t_0)\rangle - \sqrt{2}ab \langle g_z(-t_0)\rangle - b^2 \langle g_-(-t_0)\rangle \quad (3.47)$$

$$\langle g_-(t_0)\rangle = -b^{*2} \langle g_+(-t_0)\rangle - \sqrt{2}a^* b^* \langle g_z(-t_0)\rangle + a^{*2} \langle g_-(-t_0)\rangle \quad (3.48)$$

where a and b are given by equations (2.26). The transition from $+t_0$ to $+\infty$ is provided by the same diagonal transition matrix (3.45), which was already used for the transition from $-\infty$ to $-t_0$:

$$\langle g_z(+\infty)\rangle = \exp(-\theta/2) g_z(+t_0); \;\; \langle g_\pm(+\infty)\rangle = \exp(-\theta/4) g_\pm(+t_0) \quad (3.49)$$

Collecting together equations (3.45-3.49), we find the final result:

$$\langle g_z(+\infty)\rangle = \sqrt{2}e^{-3\theta/4}\left(ab^* g_+(-\infty) + a^* b g_-(-\infty)\right) + e^{-\theta}(2|a|^2-1) g_z(-\infty) \quad (3.50)$$

$$\langle g_+(+\infty)\rangle = e^{-\theta/2}\left(a^2 g_+(-\infty) - b^2 g_-(-\infty)\right) - e^{-3\theta/4}\sqrt{2}ab g_z(-\infty) \quad (3.51)$$

$$\langle g_-(+\infty)\rangle = e^{-\theta/2}\left(-b^{*2} g_+(-\infty) + a^{*2} g_-(-\infty)\right) - e^{-3\theta/4}\sqrt{2}a^* b^* g_z(-\infty) \quad (3.52)$$

Let us analyze first the case of complete decoherence at $t = -\infty$, i.e. $g_\pm(-\infty) = 0$. Then equations (3.50-3.52) look much simpler:

$$\langle g_z(+\infty)\rangle = \exp(-\theta)(2|a|^2-1) g_z(-\infty) \quad (3.53)$$

$$\langle g_+(+\infty)\rangle = -\exp(-3\theta/4)\sqrt{2}ab g_z(-\infty) \quad (3.54)$$

The equation (3.53) shows that, in the absence of decoherence the population difference can only decrease after the transition. The noise only strengthens this tendency.

However, the initial coherence if exists can increase the difference of population to the value larger than initial. The equation (3.54) demonstrates an inverse process: the coherence can appear after the transition even if it was absent in the initial state. It is straightforward to derive the transition probability from equation (3.53):

$$P_{1\to 2} = \frac{1}{2}\left[1 - \exp(-\theta)(2|a|^2 - 1)\right] = \frac{1}{2}\left(1 + \exp(-\theta)\right) - \exp(-\theta - 2\pi\gamma^2) \qquad (3.55)$$

This formula was first obtained in the work [58]. At zero noise intensity ($\theta = 0$) this result turns into the Landau-Zener transition probability. At very big noise ($\theta = \infty$) the probability is equal to 1/2. The probability is the same for transitions $1 \to 2$ and $2 \to 1$. This symmetry does not look strange keeping in mind that we considered the classical noise, which produces the induced transitions. It vanishes as soon as quantum nature of the noise and spontaneous transitions are taken into account.

Similarly the arbitrary spin problem including both regular and random transverse magnetic fields can be solved. One should employ the separation of their action in time proved in the previous section. To avoid lengthy formulas consider only the case of complete initial decoherence. Then the only non-zero components of the Bloch tensors are $g^S_{s,0}$. Their evolution is described by three independent factors, two of them originating from the noise and the central factor being the generalized Landau-Zener-Hioe matrix element:

$$g^S_{s,0}(+\infty) = \exp\left[-\frac{s(s+1)}{2}\theta\right] P_s^{0,0}(2e^{-2\pi\gamma^2} - 1) g^S_{s,0}(-\infty) \qquad (3.56)$$

where $P_s^{0,0}(x)$ is the Jacobi polynomial. The average values of the Bloch tensors components with $m \neq 0$ vanish as a result of averaging over the random phases in the initial state. To find the transition probabilities $P_{j\to j'}$ it is necessary to put all the diagonal elements of the density matrix except of ρ_{jj} equal to zero in the initial

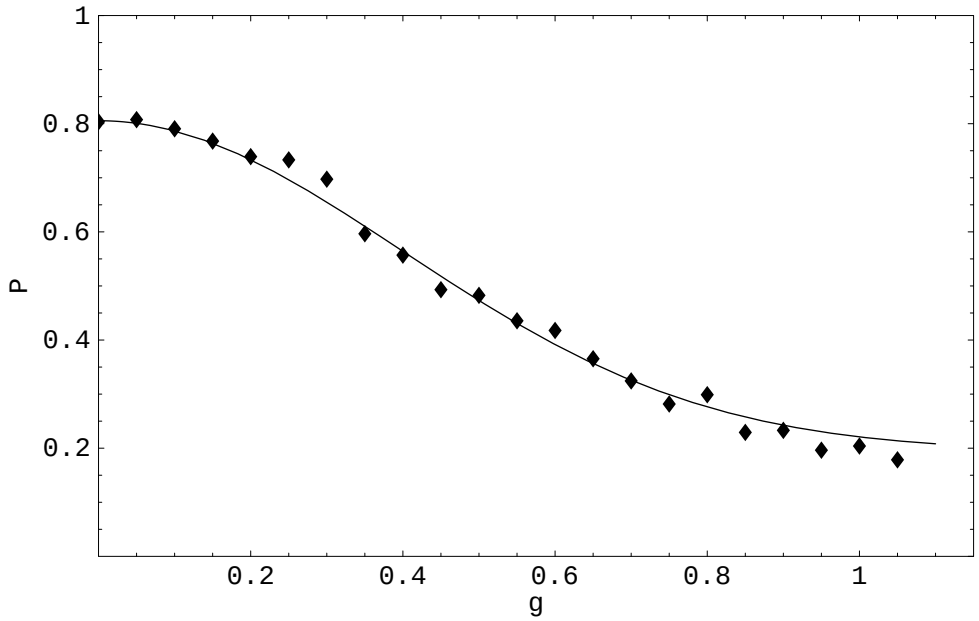

Fig. 10. The probability to stay on the same diabatic state in a two level system at constant coupling to the noise as function of the transverse magnetic field. The choice of parameters is $f_{ij} = \delta_{ix}\delta_{jx}J_x^2 e^{-\lambda|t-t'|}$, $J_x = 0.28$, $\lambda = 125$, $\beta = 1$.

state:

$$\frac{1}{2S+1} + \sum_{s=1}^{2S} g_{s,0}^S(-\infty)\left(T_{s,0}^S\right)_{k,k} = \delta_{jk}, \tag{3.57}$$

and find from these equations the initial values $g_{s,0}^S(-\infty)$. Then the transition probabilities are:

$$P_{j\to j'} = \frac{1}{2S+1} + \sum_{s=1}^{2S} g_{s,0}^S(+\infty)\left(T_{s,0}^S\right)_{j,j'} \exp\left(-\frac{s(s+1)}{2}\theta\right) \tag{3.58}$$

where $g_{s,0}^S(+\infty)$ are defined by equation (3.56).

The matching procedure, is asymptotically exact at inverse correlation time $\lambda \to \infty$. To check how it works at large but finite λ the Landau-Zener transitions subject to a fast noise were simulated numerically. The time interval of the evolution was chosen to be much larger than λ/β. The results are summarized in Fig.10 that shows

comparison of the numerically and analytically calculated probabilities to stay in the same state after the level crossing vs. the constant transverse field g at a fixed coupling to the noise $J_x = 0.28$. Each discrete point represents the averaging over 100 simulations with the same coupling constants. The solid line is the graph of the theoretical formula (2.77). One can conclude that the equation (2.77) describes well the transition probability for the LZ system subject to a fast noise.

F. Fast noise at a strong transverse regular field

The results of the previous subsection are rather general. They are valid for arbitrary strength of noise and arbitrary form of noise correlators. However two main assumptions were set in derivation.

First, although noise was allowed to be anisotropic, the relative strengths of noise in transverse and longitudinal directions were supposed to be of the same order of magnitude. As a result, the diagonal component of random field was proved to be not important since diagonal noise does not couple diabatic states directly. This may not be true in some applications where nondiagonal noise is strongly suppressed for some reasons in comparison to diagonal one.

Second, the time scales of noise accumulation and transitions due to regular transverse field were supposed to be well separated. This may be no longer true in the case of extremely strong regular transverse field. Without noise this case would correspond to adiabatic limit with transition probability almost equal to unity. In this section the solution in such an adiabatic limit will be shown and the following section is devoted to the problem of strong diagonal noise.

Consider the situation of adiabatically changing levels, i.e. $\dot{b}_z \ll b_x^2$, and the noise is supposed to be sufficiently fast $\tau_n \ll (\dot{b}_z)^{-1/2}$. We will not specify the relationship

between b_x and τ_n. In experiments with nanomagnets the inequality $\sqrt{\dot{b}_z}\tau_n \ll 1$ (fast noise condition) can be easily realized since the sweeping rate of the applied magnetic field can be made arbitrarily small. However, the noise may be not fast enough to compete with the regular part of the tunneling amplitude b_x. The nuclear bath correlation time is in the range of $\tau_n \sim 1ms$, whereas the measured values of the tunneling amplitude for known nanomagnets range between $10^{-10} - 10^{-3}K$, or equivalently $10 - 10^8 Hz$. In a part of this interval $b_x\tau_n$ has a rather large value so that time scale separation employed in the previous section does not take place. Fortunately in such a limit of a strong regular part of the transverse field other simplifications are possible. We consider in this section only 2-level systems. A natural approach to this problem is the transfer to the adiabatic set of states, i.e. to the eigenstates of the instantaneous regular part of the Hamiltonian (3.5). Lets denote this time-dependent eigenvectors as $a(t) = \begin{pmatrix} a_1(t) \\ a_2(t) \end{pmatrix}$ and $b(t) = \begin{pmatrix} -a_2(t) \\ a_1(t) \end{pmatrix}$, where $a_1(t) = \sqrt{\frac{\varepsilon(t)+b_x}{2\varepsilon}}$; $a_2(t) = \sqrt{\frac{\varepsilon(t)-b_x}{2\varepsilon}}$ and $\varepsilon(t) = \sqrt{b_z^2 + b_x^2}$ ($b_z = \dot{b}_z t$). The unitary matrix of transformation to the adiabatic set reads:

$$U(t) = \begin{pmatrix} a_1(t) & a_2(t) \\ -a_2(t) & a_1(t) \end{pmatrix} = a_1(t)I - ia_2(t)\sigma_y \tag{3.59}$$

where σ_α ($\alpha = x, y, z$) are Pauli matrices. In the new basis the total Hamiltonian acquires the following form:

$$H = \varepsilon(t)\sigma_z + U\eta\sigma U^{-1} = \varepsilon(t)\sigma_z + \eta'\sigma \tag{3.60}$$

The transformation of the random field components is:

$$\eta'_x = (a_1^2 - a_2^2)\eta_x + 2a_1a_2\eta_z = \frac{b_z}{\varepsilon}\eta_x + \frac{b_x}{\varepsilon}\eta_z;\ \eta'_y = \eta_y;\ \eta'_z = -\frac{b_x}{\varepsilon}\eta_x + \frac{b_z}{\varepsilon}\eta_z \qquad (3.61)$$

In this form the Hamiltonian (3.60) essentially coincides with the Hamiltonian of the auxiliary problem (zero regular transverse field) for the two-level system (see Section B). The essential difference is first, that the effective regular external field is not linear in time; instead it is equal to $\varepsilon(t)$; second, the correlators of effective noise η' now depend not only on the time difference, but also on time itself due to the time-dependent transformation (3.61):

$$\begin{aligned} \langle \eta'_x(t)\eta'_x(t')\rangle &= \frac{1}{\varepsilon(t)\varepsilon(t')}\left[\dot{b}_z^2 tt' \langle \eta_x(t)\eta_x(t')\rangle + b_x^2 \langle \eta_z(t)\eta_z(t')\rangle + \right. && (3.62)\\ &\left. \dot{b}_z b_x t \langle \eta_x(t)\eta_z(t')\rangle + \dot{b}_z b_x t' \langle \eta_z(t)\eta_x(t')\rangle\right] && (3.63) \end{aligned}$$

Still the noise correlation time is small in comparison to the characteristic time of variation for the adiabatic energy $\varepsilon(t)$. Note also that since the adiabatic basis is time-dependent, there are should be additional terms in evolution equations due to nonadiabatic corrections. However we can disregard them if regular transverse field is strong, i.e. in the case that we consider. Employing the same approximation as in Section B, we arrive at a similar equation of motion for averaged Bloch vector components in adiabatic basis in the case of complete initial decoherence:

$$\langle \dot{g}'_z(t)\rangle = -F'(t)\langle g'_z(t)\rangle \qquad (3.64)$$

where

$$\begin{aligned} F'(t) &= \hat{f}_{yy}(\varepsilon(t)) + \frac{1}{\varepsilon^2(t)}\left[\dot{b}_z^2 t^2 \hat{f}_{xx}(\varepsilon(t)) + b_x^2 \hat{f}_{zz}(\varepsilon(t)) \right. && (3.65)\\ &\left. + \dot{b}_z b_x t\left(\hat{f}_{xz}(\varepsilon(t)) + \hat{f}_{zx}(\varepsilon(t))\right)\right] && (3.66) \end{aligned}$$

In the last equation the hats symbolize Fourier-transforms of corresponding correlators. As before, we can find the average value $\langle g_z(t)\rangle$ at arbitrary moment of time. Asymptotically at $t \to +\infty$ we find:

$$\langle g_z'(+\infty)\rangle = \exp\left[-\int\limits_{-\infty}^{\infty} \frac{\dot{b}_z^2 t^2 \hat{f}_{xx}\left(\varepsilon(t)\right) + \varepsilon^2(t)\hat{f}_{yy}\left(\varepsilon(t)\right) + b_x^2 \hat{f}_{zz}\left(\varepsilon(t)\right)}{\varepsilon^2(t)} dt\right] \langle g_z'(-\infty)\rangle \tag{3.67}$$

The characteristic time after which the correlators in equation (3.67) become very small and decay rapidly is determined by approximate equation $\varepsilon(t)\tau_n \approx 1$. If $\tau_n \ll b_x^{-1}$, then this characteristic time coincides with the accumulation time $t_{acc} = \left(\dot{b}_z \tau_n\right)^{-1}$, terms proportional to $b_x$are negligibly small and we return to the result (3.50) with $|a| = 1$, or equivalently to (3.28). In the opposite case $\tau_n \gg b_x^{-1}$ the value $\varepsilon(t)$ exceeds τ_n^{-1} at any moment of time t. Therefore, all correlators are small and the value of exponent in (3.67) is close to 1. It means that practically no transition proceeds due to the noise between adiabatic states. Thus, equation (3.67) carries most interesting information when $\tau_n \sim b_x^{-1}$.

Lets examine the formula (3.67) in two most typical cases namely of diagonal and of isotropic noise. In what follows we will assume exponentially correlated noise in two state system with the Hamiltonian.

$$\hat{H} = \beta t\hat{\sigma}_z + g\hat{\sigma}_x + \sum_{i=x,y,z} \eta_i \hat{\sigma}_i, \quad (\langle \eta_i(t)\eta_j(t'\rangle = f_{ij}(t-t')) \tag{3.68}$$

1) For isotropic noise $f_{ij}(t-t') = \delta_{ij} J^2 e^{-\lambda|t-t'|}$

one finds:

$$\langle g_z'(+\infty)\rangle_{noise} = \exp\left(-\frac{4\pi J^2}{\beta}\frac{\lambda}{\sqrt{\lambda^2+g^2}}\right) \tag{3.69}$$

This formula is derived in the limit $g \gg \beta^{1/2}$. In the limit also $\lambda \gg g$ it coincides with predictions of the result (3.55) that is also valid in this limit. Thus the formula (3.69) generalizes that result to the case of arbitrary magnitude of the parameter g.

2)Assume now the noise is only along the sweeping field (as it is often the case in molecular magnets) i.e. $f_{ij}(t-t') = \delta_{iz}\delta_{jz}J^2 e^{-\lambda|t-t'|}$. Then $F(|t_1-t_2|) = \frac{g^2J^2}{g^2+(\beta t)^2}$ and

$$\langle g'_z(+\infty)\rangle_{noise} = \exp\left(-\frac{2\pi J^2}{\beta}\frac{(\lambda/g)}{1+(\lambda/g)^2+\sqrt{1+(\lambda/g)^2}}\right) \tag{3.70}$$

In the limits $\lambda \gg g$ and $\lambda \ll g$ we find $g'_z(+\infty) = 1$ that means the system remains on the same adiabatic state. The same result would be in the absence of noise. The transition probability becomes sensitive to diagonal noise only when $\lambda \sim g$ or in the case of very strong noise $\frac{J^2}{\beta} \gg 1$.

An interesting feature of the transition probability is that the z-component of noise can produce transitions between adiabatic states. This happens because the latter rotates with time. Note that z-component of noise is irrelevant if $\tau_n \ll b_x^{-1}$.

G. Strong diagonal noise

The non-diagonal δ-like white noise leads to the equilibration of population between all states. The effect of a white noise directed along the sweeping field is different. Such a noise does not couple different states but its action leads to the loss of coherence. As it was shown earlier, in the case of a 2-level system it results in a Debye-Waller factor for ρ.

1. Arbitrary spin in a strong diagonal random field

Consider the case of a general spin S placed into a regular field $\mathbf{h}_0 = \hat{z}\beta t + \hat{x}\Gamma$ and the random field directed along z-axis. Its Hamiltonian reads:

$$\hat{H}_{tot} = \beta t\hat{S}_z + \Gamma\hat{S}_x + \eta(t)\hat{S}_z \tag{3.71}$$

Assume $< \eta(t)\eta(t') >= 2\gamma\delta(t-t')$.

The master equation for averaged density matrix elements can be derived in similar way as the one for nondiagonal noise. One should expand the solution of equation (3.71) in the power series over the noise amplitude and average each term. The resulting series in powers of γ is a formal solution of a differential equation known as a master equation. For the coupling to diagonal noise it has been known for long time [49]. In case of the Hamiltonian (3.71) the master equation has the following form:

$$\frac{\partial\hat{\rho}(t)}{\partial t} = -i[\hat{H}(t),\hat{\rho}(t)] - \gamma[\hat{S}_z,[\hat{S}_z,\hat{\rho}(t)]] = \\ -i[\beta t\hat{S}_z + \Gamma\hat{S}_x,\hat{\rho}(t)] - \gamma(\hat{S}_z^2\hat{\rho}(t) + \hat{\rho}(t)\hat{S}_z^2 - 2\hat{S}_z\hat{\rho}\hat{S}_z) \tag{3.72}$$

It is convenient to introduce notations $\Gamma_{ij} = \Gamma < i|S_x|j >$. The equations for diagonal

and non-diagonal elements read:

$$\begin{aligned} &\dot{\rho}_{ii} = -i(\Gamma_{i,i-1}(\rho_{i-1,i} - \rho_{i,i-1}) + \Gamma_{i,i+1}(\rho_{i+1,i} - \rho_{i,i+1})) \\ &i = -S, -S+1, \ldots, S \end{aligned} \tag{3.73}$$

$$\begin{aligned} &\dot{\rho}_{mn} = -(i\beta t(m-n) + \gamma(n-m)^2)\rho_{mn} - \\ &-i(\Gamma_{m,m-1}\rho_{m-1,n} - \rho_{m,n+1}\Gamma_{n+1,n} + \Gamma_{m,m+1}\rho_{m+1,n} - \rho_{m,n-1}\Gamma_{n-1,n}), \;\; m \neq n \end{aligned} \tag{3.74}$$

It is possible to find the asymptotically exact solution of equations (3.73), (3.74) in the limit of strong noise $\gamma \gg \Gamma, \beta$. In this limiting case, non-diagonal elements of the density matrix are $\sim \Gamma/\gamma$ times smaller than diagonal ones. Indeed, lets disregard the dynamical term $\dot{\rho}_{ij}$ in equations for the non-diagonal elements. Then equation (3.74) at $n = m \pm 1$ implies that the non-diagonal elements $\rho_{n,n\pm1}$ are suppressed comparing with diagonal matrix elements by the factor $\sim \Gamma/\gamma$. The matrix elements $\rho_{n,n\pm2}$ are suppressed by the same factor with respect to $\rho_{n,n\pm1}$ etc. The characteristic time interval following from equation (3.73) is $\Delta t \sim |\rho_{nn}/(\Gamma\rho_{n,n\pm1})| \sim \gamma/\Gamma^2$. From this estimate we find that the time derivative of the largest non-diagonal matrix element $|\dot{\rho}_{i,i\pm1}| \sim (\Gamma^2/\gamma)|\rho_{i,i\pm1}| \ll \gamma\rho_{i,i\pm1}$ can be neglected. Retaining only main diagonal and two adjacent non-diagonals in the matrix equations (3.73),(3.74), the non-diagonal elements in terms of diagonal read:

$$\begin{aligned} &\rho_{i+1,i} = -\tfrac{i\Gamma_{i+1,i}}{i\beta t+\gamma}(\rho_{i,i} - \rho_{i+1,i+1}), \\ &\rho_{i,i+1} = \tfrac{i\Gamma_{i,i+1}}{i\beta t-\gamma}(\rho_{i+1,i+1} - \rho_{i,i}), \\ &\rho_{i,j} = 0, \;\; |i-j| > 1 \end{aligned} \tag{3.75}$$

The problem is reduced to determining of the $2S+1$ diagonal elements. Lets introduce a vector $\mathbf{c}$ with the coordinates $c_i = \rho_{ii}$. They are probabilities to find the spin in a particular eigenstate of the operator S_z. Substitution of (3.75) into (3.73) gives a

differential equation for the vector $\mathbf{c}(t)$

$$\dot{\mathbf{c}}(t) = (\frac{\Gamma^2}{i\beta t + \gamma} - \frac{\Gamma^2}{i\beta t - \gamma})\hat{M}\mathbf{c}(t) \tag{3.76}$$

were the constant matrix $\hat{M}$ has following matrix elements:

$$\begin{aligned} M_{ii} &= -(< i+1|\hat{S}_x|i >^2 + < i|\hat{S}_x|i-1 >^2) = -\tfrac{1}{2}(S^2 + S - i^2) \\ M_{i,i+1} &= M_{i+1,i} =< i+1|\hat{S}_x|i >^2 = \tfrac{1}{4}(S+i+1)(S-i) \end{aligned} \tag{3.77}$$

All other elements are zeros. Equation (3.76) can be easily integrated

$$\mathbf{c}(t) = \exp(\int_{t_0}^{t} \frac{2\gamma\Gamma^2}{(\beta t')^2 + \gamma^2} dt' \hat{M})\mathbf{c}(t_0) \tag{3.78}$$

To find transition probabilities one should take limits of integral in equation (3.78) as $t_0 = -\infty$ and $t = +\infty$:

$$\mathbf{c}(+\infty) = \exp(\frac{2\pi\Gamma^2}{\beta}\hat{M})\mathbf{c}(-\infty) \tag{3.79}$$

This result demonstrates that, for a general spin S as well as for a spin 1/2, the transition probabilities do not depend on the specific value of γ provided that γ is large. Below the transition probabilities for some values of spin are explicitly written. Denote $E_1 = e^{-\Gamma^2\pi/\beta}$, $E_2 = e^{-3\Gamma^2\pi/\beta}$ and $E_3 = e^{-6\pi\Gamma^2/\beta}$. Then for $S = 1/2$ the formula (3.79) reads

$$\begin{aligned} P_{1/2\to 1/2} &= P_{-1/2\to -1/2} = \tfrac{1}{2}(1 + E_1) \\ P_{1/2\to -1/2} &= P_{-1/2\to 1/2} = \tfrac{1}{2}(1 - E_1) \end{aligned} \tag{3.80}$$

For $S = 1$

$$\begin{aligned} P_{1\to 1} &= P_{-1\to -1} = \tfrac{1}{6}(2 + E_2 + 3E_1) \\ P_{1\to 0} &= P_{-1\to 0} = P_{0\to 1} = P_{0\to -1} = \tfrac{1}{3}(1 - E_2) \\ P_{1\to -1} &= P_{-1\to 1} = \tfrac{1}{6}(2 + E_2 - 3E_1) \\ P_{0\to 0} &= \tfrac{1}{3}(1 + 2E_2) \end{aligned} \tag{3.81}$$

The results (3.80) and (3.81) coincide with known solutions for two- and three-level LZ models with strong decoherence [49]. The formula (3.80) for two levels was first derived by Kayanuma [46] and in the literature is often called the Kayanuma formula. A new result for a spin $S = 3/2$ that follows from (3.79) reads:

$$\begin{aligned}
&P_{3/2\to 3/2} = P_{-3/2\to -3/2} = \tfrac{1}{4} + \tfrac{1}{20}E_3 + \tfrac{1}{4}E_2 + \tfrac{9}{20}E_1 \\
&P_{3/2\to 1/2} = P_{-3/2\to -1/2} = P_{1/2\to 3/2} = P_{-1/2\to -3/2} = \tfrac{1}{4} - \tfrac{3}{20}E_3 - \tfrac{1}{4}E_2 + \tfrac{3}{20}E_1 \\
&P_{3/2\to -1/2} = P_{-3/2\to 1/2} = P_{-1/2\to 3/2} = P_{1/2\to -3/2} = \tfrac{1}{4} + \tfrac{3}{20}E_3 - \tfrac{1}{4}E_2 - \tfrac{3}{20}E_1 \\
&P_{3/2\to -3/2} = P_{-3/2\to 3/2} = \tfrac{1}{4} - \tfrac{1}{20}E_3 + \tfrac{1}{4}E_2 - \tfrac{9}{20}E_1 \\
&P_{1/2\to 1/2} = P_{-1/2\to -1/2} = \tfrac{1}{4} + \tfrac{9}{20}E_3 + \tfrac{1}{4}E_2 + \tfrac{1}{20}E_1 \\
&P_{1/2\to -1/2} = P_{-1/2\to 1/2} = \tfrac{1}{4} - \tfrac{9}{20}E_3 + \tfrac{1}{4}E_2 - \tfrac{1}{20}E_1
\end{aligned} \tag{3.82}$$

In adiabatic limit $\Gamma^2/\beta >> 1$ all states are equally populated after the evolution.

If measurements of a Landau-Zener transition probability are performed on a small number of samples then the result will fluctuate since the amplitude now is a random variable that depends on a noise realization. Using results for higher spins it is possible to estimate the mean square deviation of the transition probability from its average in a single two level system. For the same time dependence of external fields the nonaveraged over noise transition probabilities for spin 1/2 and 1 are related by $P_{1\to 1} = P^2_{1/2\to 1/2}$ [39]. Applying expressions from (3.80) and (3.81) gives:

$$< \Delta P^2_{1/2\to 1/2} > = < P^2_{1/2\to 1/2} > - < P_{1/2\to 1/2} >^2 = \frac{1}{12} + \frac{E_2 - E_1}{6} - \frac{E_1^2}{4} \tag{3.83}$$

2. Electron motion driven by electric field

The muster equation in the strong decoherence limit can be easily studied in another multistate Landau-Zener model namely in the model of electron transitions in a linear chain of sites driven by an external field. The noise in such a system arises due to thermal fluctuations chaotically changing the energy of the electron.

Suppose that there are no correlations of noise at different sites and denote $|n\rangle$ a state located at the n-th site of the chain. In terms of this set the electron Hamiltonian with linear dependence of an external field on time reads:

$$\hat{H} = \sum_n (g \mid n\rangle\langle n+1 \mid + c.c.) + nvt \mid n\rangle\langle n \mid + \eta_n(t) \mid n\rangle\langle n \mid \tag{3.84}$$

where v and g are constants and it is assumed that the noise power is the same for all sites i.e. $< \eta_m(t)\eta_n(t') >= 2\gamma\delta_{mn}\delta(t-t')$. Here only the limit of strong noise $\gamma >> g$ will be considered. Then, as in previous example, the non-diagonal elements of the density matrix $\rho_{i,j}$ with $|i-j| > 1$ can be neglected. Equations for diagonal matrix elements of the density matrix are:

$$\dot{\rho}_{nn} = -ig(\rho_{n+1,n} + \rho_{n-1,n} - \rho_{n,n+1} - \rho_{n,n-1}) \tag{3.85}$$

Equations for non-diagonal elements after the averaging over the random noise read:

$$\begin{aligned} \dot{\rho}_{n+1,n} &= (-ivt - 2\gamma)\rho_{n+1,n} - ig(\rho_{n,n} - \rho_{n+1,n+1}) \\ \dot{\rho}_{n-1,n} &= (ivt - 2\gamma)\rho_{n-1,n} - ig(\rho_{n,n} - \rho_{n-1,n-1}) \end{aligned} \tag{3.86}$$

Neglecting again time derivatives in these equations, one can find:

$$\begin{aligned} \rho_{n+1,n} &= \tfrac{-ig}{ivt+2\gamma}(\rho_{n,n} - \rho_{n+1,n+1}) \\ \rho_{n-1,n} &= \tfrac{ig}{ivt-2\gamma}(\rho_{n,n} - \rho_{n-1,n-1}) \end{aligned} \tag{3.87}$$

Substituting (3.87) into the equations (3.85) for diagonal elements, one obtains the evolution equations for diagonal matrix elements of the density matrix $\rho_{n,n}$:

$$\dot{\rho}_{n,n} = g^2 \left(\frac{1}{ivt - 2\gamma} - \frac{1}{ivt + 2\gamma} \right) (2\rho_{n,n} - \rho_{n+1,n} - \rho_{n-1,n}) \tag{3.88}$$

Without loss of generality one can assume that initially, at $t = -\infty$, the particle was located at the site number zero. It means that the initial conditions for the

master equation are $\rho_{0,0}(t=-\infty)=1$ and all other elements of the density matrix are zeros at $t=-\infty$. Then diagonal matrix elements $\rho_{n,n}$ acquire the meaning of transition probabilities from zeroth to the n-th site at a current time t. As in the previous example we can find the solution for a chain of arbitrary number of sites in the matrix form.

In the limit of infinite number of sites a compact solution can be found by employing the Fourier-transformation $\rho_{n,n}=\frac{1}{2\pi}\int_0^{2\pi}e^{in\phi}u(\phi,t)$. The system of coupled differential equations (3.88) is diagonalized by this transformation. Corresponding differential equation of the first order for the function $u(\phi,t)$ is readily solved. Its solution with the initial condition $u(\phi,-\infty=1)$ is

$$u(\phi,t)=\exp\left[-2\left(\frac{\pi}{2}+\arctan\frac{vt}{2\gamma}\right)(1-\cos\phi)\right] \tag{3.89}$$

In the limit $t\to+\infty$ it approaches its limiting value:

$$u(\phi,t\to+\infty)=e^{\frac{-4\pi g^2}{v}(1-\cos\phi)} \tag{3.90}$$

After the inverse Fourier-transformation the diagonal elements of the density matrix are given by

$$\rho_{n,n}(t\to+\infty)=e^{-\frac{4\pi g^2}{v}}I_n(\frac{4\pi g^2}{v}) \tag{3.91}$$

As we already said $\rho_{n,n}$ is the transition probability from the site with the index 0 to the cite with the index n .

It is interesting to compare results of this calculation which incorporates a strong noise with the transition probabilities without noise. In the absence of the noise ($\gamma=0$) the transition probabilities are [41]:

$$P_n^{(coh)}=|J_n(\sqrt{8\pi}g)|^2 \tag{3.92}$$

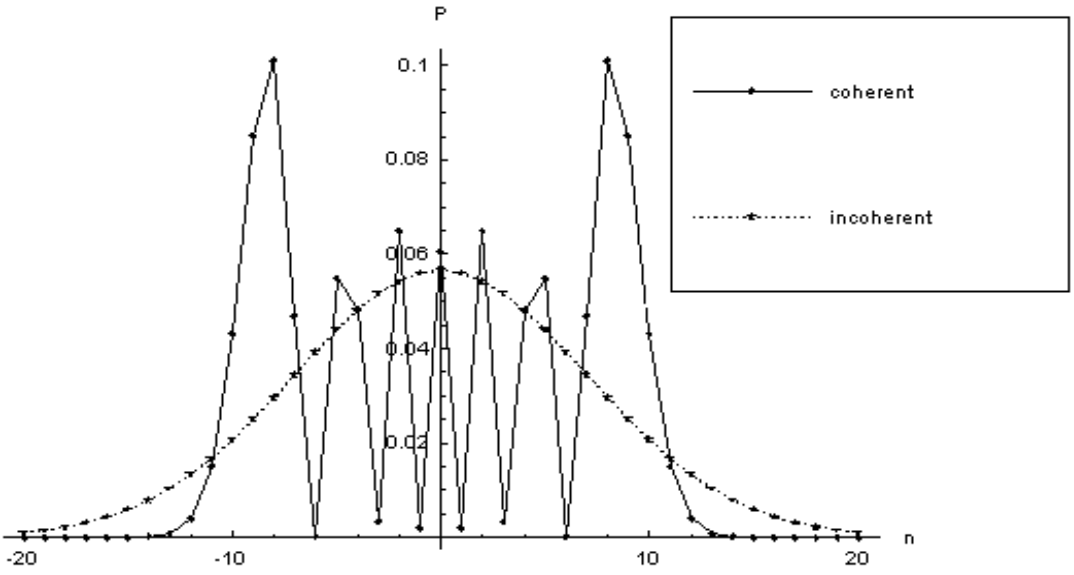

Fig. 11. Comparison of transition probabilities in coherent (without noise) and incoherent (with strong noise) LZ models on the chain.

Fig.11 shows a typical behavior of the transition probabilities for both cases. The difference in the behavior is clearly pronounced. In the absence of the noise the transition probabilities oscillate as functions of n and g (see [41] for details). These oscillations arise due to the interference among the amplitudes of different Feynman paths leading from the initial to the final point. In the case of strong noise these oscillation are suppressed by the noise imposed decoherence and the probability distribution is a smooth bell-like curve. A simple parameter that is related to the effective diffusion coefficient and can be measured experimentally is the average square displacement of the particle during one sweep of the external field. For a chain with a strong noise it is:

$$< n^2 >= \sum_{n=-\infty}^{+\infty} n^2 e^{\frac{-4\pi g^2}{v}} I_n(\frac{4\pi g^2}{v}) = \frac{4\pi g^2}{v} \tag{3.93}$$

Despite a strong difference in the distribution functions, the average square displacement (3.93) coincides with that for the coherent evolution without noise.

CHAPTER IV

SPIN-BATH EFFECTS ON LANDAU-ZENER TRANSITIONS IN MOLECULAR NANOMAGNETS

Magnetic molecules constitute ideal prototype systems for investigating experimentally basic physical phenomena at nanoscale, such as tunneling of a large collective electronic spin interacting with its environment [103, 104, 105, 7, 106]. A substantial contributions to the theory of spin tunneling in these molecules was made by theorists [59, 107, 108, 109, 110]. Understanding of these phenomena is crucial for fundamental science as well as for possible applications. In particular, extensive analytical work has been done in order to understand the effect of the nuclear spin bath decoherence on the tunneling of a large spin [58, 59, 108], although some issues still exist [109]. Several predictions of this theory have been confirmed in experiments on magnetic relaxation in the crystals of magnetic molecules [103] subjected to a constant external magnetic field ("hole digging" regime). It has been demonstrated that by changing the number of nuclear spins per molecule by using isotopic substitution, the relaxation rate can change considerably (up to two orders of magnitude). The similar, but much weaker isotopic effect was observed in the experiments on the spin tunneling in a linearly varying magnetic field passing the avoided level crossing, the Landau-Zener transition.

For our purposes, the magnetic state of the molecule can be described as a large single spin ($S = 10$ for both Mn_{12} and Fe_8). Due to large easy-axis anisotropy, the two lowest states $S_z = \pm S$ are nearly degenerate and separated from the others by a large gap (14.4 K for Mn_{12}, 5.5 K for Fe_8). Due to tunneling which mixes the states $S_z = +S$ and $S_z = -S$, the two lowest states are split by a very small tunneling amplitude Δ. At low temperatures, when the thermally activated population of

higher states can be neglected, tunneling is the only mechanism for magnetization relaxation. At low-temperature it is sufficient to project the full spin Hamiltonian of the molecule onto the lowest two states and take into account the interaction of the electronic spin with the bath of nuclear spins present in the molecule. The influence of the spin-phonon interactions is not important at low temperatures. Moreover, the dipolar interactions between the electronic spins of different molecules are noticeably strong, and should be taken into account, at least as some external magnetic field, which might change with time.

In [58, 59, 111], the above-mentioned approach was employed, resulting in the following Hamiltonian for the effective two-state model for the electronic spin:

$$\hat{H} = \Delta\hat{\tau}_x + \xi_e(t)\hat{\tau}_z + \sum_{i=1}^{N} \omega_{||}^i \hat{\sigma}_z^i \hat{\tau}_z + \sum_{i=1}^{N} \vec{\omega}_{\perp}^i \hat{\vec{\sigma}}^i + \sum_{i<j=1}^{N} V_{ij}^{\alpha\beta} \hat{\sigma}_\alpha^i \hat{\sigma}_\beta^j \tag{4.1}$$

where the operators $\hat{\tau}_{x,y,z}$ are the Pauli operators acting on the central two level system, and $\hat{\sigma}_{x,y,z}^i$ are the Pauli operators acting on the i-th nuclear spin. The first two terms describe, correspondingly, the tunneling of the central spin, and the Zeeman energy of this spin in the external field ξ_e. The third term describes the bias produced on the central spin by the nuclear spins due to the hyperfine interaction of the central and nuclear spins. The parameters $\omega_{||}^i$ are the strengths of hyperfine couplings to corresponding nuclear spins. Index i enumerates nuclear spins and Greek indexes enumerate the spin projections x, y, and z. N is the number of the nuclear spins in a molecule, typically $N \sim 10^2$. The fourth term describes interaction of nuclear spins with external fields, e.g. dipolar fields in the sample. In the literature it is often called the "orthogonality blocking" term. This term is responsible for the co-flips of the nuclear spins and the central spin; such co-flips increase the orthogonality of the initial and final states of the spin bath, thus decreasing the probability of the central spin flip (i.e. blocking the central spin in its initial state) at a given resonance.

Its second effect is the appearance of new tunneling resonances between states with distinct states of the spin bath. The sum of the third and the fourth terms may be viewed as the total field acting on the nuclear spin k, either $\vec{\omega}_{||}^k + \vec{\omega}_{\perp}^k$ or $-\vec{\omega}_{||}^k + \vec{\omega}_{\perp}^k$ depending on the orientation of the central spin, and the $\vec{\omega}_{||}^k$ component describes coupling between the two subsystems (the direction of $\vec{\omega}_{||}^k$ is chosen to be the $\hat{z}$-axis for the nuclear spin k). According to [58, 59], the orthogonality blocking mechanism is characterized by a parameter $\kappa = (1/2)\sum_i(\omega_{\perp}^i/\omega_{||}^i)^2$. For $\omega_{\perp}^i \ll \omega_{||}^i$, the value of κ represents the typical number of nuclear spin co-flips occurring during a tunneling event of the electronic spin of a nanomagnet. Up to now, most of the analytical results belong to the situation when κ is not large. However, for consideration of real magnetic molecules, such as Fe_8, the case of $\omega_{\perp}^i \sim \omega_{||}^i$ is appropriate, since nanomagnets have a wide distribution of the hyperfine coupling constants $\omega_{||}^i$, and the parameters $\omega_{\perp}^i$ may not be considered as small in comparison with $\omega_{||}^i$ [112]. The last term, which is called the spin-diffusion term, describes the dipolar interaction of the nuclear spins with each other. In real nanomagnets $V_{ij}^{\alpha\beta} \sim 10^{-7} - 10^{-9}$ K, that is much smaller than the hyperfine interaction $\omega_{||}^i \sim 10^{-4}$ K, but $V_{ij}^{\alpha\beta}$ still can be larger than the tunneling amplitude Δ. For example, $\Delta \sim 10^{-8}$ K in Fe_8 molecules, and $\Delta \sim 10^{-10}$ K in Mn_{12}. Moreover, dipolar interactions between the nuclear spins lead to chaotic evolution of the bath and hence can strongly enhance the relaxation [58].

Because of the difference in the nuclear, and electronic magnetic moments, $\mu_N/\mu_S \sim 10^{-4}$, we ignore the time dependent component of the external field acting on nuclear spins because when $\mu_N B(t) \sim \omega_k$ we have $\mu_S B(t)$ much larger than all the other energy scales, and the electronic spin system is already frozen in the semiclassical up or down state. In (4.1) the terms proportional to $\hat{\tau}_x\hat{\vec{\sigma}}$ accounting for the tunneling amplitude dependence on the field generated by nuclear spins are not included. Their effect is proportional to ω_k/K, where K is the anisotropy field, and for the

Fe_8 molecule ω_k/K is estimated to be below 1% while the isotope effect observed in [103] is about 50%. [Similarly, the dependence of Δ on transverse components of $\mathbf{B}_{dip}$ results in a relatively narrow (about 10%) distribution of tunneling amplitudes, which may be easily accounted for by averaging the final answers].

Dynamics of the electronic spin in magnetic molecules, as described by the Hamiltonian (4.1), has been analyzed in a number of works [58, 59, 111, 113] suggesting the following qualitative picture. First, this dynamics is determined by the dipolar interactions between the bath spins (spin diffusion term in Eq. 4.1), Since the value of Δ is small, the spin tunneling is allowed only if the two corresponding levels are separated by energy of order of Δ or even less if orthogonality blocking is strong. The separation of these levels is determined by the bias field ξ_e and the effective field produced by the nuclear spins. The latter field fluctuates due to the flips of the bath spins and hence can make quantum levels pass through the resonances; therefore the effective "window" of external static fields that allow the relaxation becomes much wider i. e. of the order of the amplitude of such hyperfine field fluctuations. Second, the dynamics of the nuclear spins is determined by their coupling to the central spin. The theory [113] predicts that for $\kappa \sim 1$ the effective window of external biases resulting to a fast relaxation grows approximately as $\xi \sim \kappa\xi_0$ and for $\kappa \sim O(N^{1/2})$ or higher it becomes of the same order [112] as the half width of the whole nuclear spin multiplet E_0, where $E_0^2 \sim \sum_{i=1}^{N}(\omega_{||}^i)^2$. The orthogonality blocking mechanism suppresses tunneling at a given resonance, however the possibility of nuclear spin co-flips allows tunneling between resonances with different final states of the bath.

Measurements of the hysteresis loop at low temperature in molecular nanomagnets reveal the discrete steps of magnetization which were interpreted as the sequence of LZ transitions between the central spin quantum levels [114]. After this finding, the Landau-Zener theory was employed in the experimental technique that allows

to measure tunneling parameters of the effective spin Hamiltonian for nanomagnets [105]. In such measurements the Zeeman coupling of the central spin S with magnetic moment μ_S was made linearly time dependent by coupling central spin to sweeping magnetic field, i.e. $\xi_e(t) = \beta t \equiv \mu_S B(t)$. Any static component of the external field then may be compensated by the proper selection of the time origin if the amplitude of the sweep is large enough. In molecular magnets, electronic spins of different molecules couple to each other by dipole forces which are equivalent to quasi-static fields, B_{dip}, evolving along with the sample-shape dependent magnetization distribution [58]. This effect, and the *collective* relaxation in an ensemble of electronic spins is important only in the limit of small sweeping rates not considered here. For small amplitude sweeps ($B_{max} < B_{dip}$) the result of the measurement necessarily involves the fraction of molecules which have a chance to relax at the avoided crossing and may not be analyzed using equations (4.2) and (4.3). In what follows it is assumed that $B_{max} \gg B_{dip}$. By measuring transition rates in external time-dependent magnetic field, the tunneling splitting, Δ, can be deduced from the Landau-Zener formula

$$P = 1 - e^{-\pi\Delta^2/\beta} \,, \tag{4.2}$$

where β is the sweeping rate of the external field, and P is the transition probability per level crossing in a system described by the Hamiltonian $H_{LZ} = \xi_e(t)\,\hat{\tau}_z + \Delta\,\hat{\tau}_x$ with the time-dependent external bias field $\xi_e(t) = \beta t$.

At very low temperature the dynamics of the magnetization in nanoparticles is strongly influenced by nuclear spins, and predictions based on the spin bath theory were recently verified experimentally [103, 7, 106]. In the limit of vanishingly small Δ, relevant for the Fe_8 and Mn_{12} molecular magnets, the hyperfine, transfer hyperfine and dipole interactions between the electronic and nuclear spins render the two-level dynamics of the central spin incoherent. This result questions the limits of the validity

of the LZ formula, and, correspondingly, under what conditions it can be employed for measurements of the tunneling energy splitting in nanomagnets. On one hand, for very fast sweeping rates the result

$$P\beta = \pi\Delta^2 \tag{4.3}$$

is universally valid irrespective of the coupling to any environment with finite energy spectrum. Indeed, if ω_c is the largest energy scale characterizing the spectrum of the system and environment (with or without coupling), then for $\beta \gg \omega_c^2$ we are in the limit of the sudden perturbation theory which predicts that corrections to Eq. (4.3) are small at least in the parameter ω_c^2/β. However, when coupling to the environment is strong, one may not exclude the possibility of having another plateau in $P\beta$ at intermediate sweeping rates corresponding, e.g. to the "renormalized" tunneling splitting.

Sinitsyn and Prokof'ev [61] analyzed analytically the LZ transitions in a system describing a central spin coupled to the spin bath starting from the microscopic Hamiltonian (4.1); the numerical simulations of the problem were performed by Sinitsyn and Dobrovitsky [62].

A. Effect of the spin diffusion

The spin bath Hamiltonian (last term in Eq. (4.1) includes interactions among nuclear spins with a typical energy scale of order $10^{-8} \div 10^{-7} K$. This term gives dynamics to the nuclear spins (spin diffusion) and makes the "internal" bias field $\xi_N = \sum_N \hat{\sigma}_z^k \vec{\omega}_k$ fluctuate in time. It is clear from Eq. (4.1), that fluctuating ξ_N helps *incoherent* relaxation if ξ_e is static and out of resonance $\xi_e >> \Delta$ by creating an effective resonance window of width $\xi_0 \gg \Delta$ determined by the root mean square parameter,

$\xi_0 = \sqrt{\langle \xi_N^2(t) \rangle}$, where $\langle \ldots \rangle$ stands for the averaging over random processes. Under the assumption that all environmental states may be sampled in the course of nuclear spin diffusion/relaxation we have $\xi_0^2 = \sum_{k=1} \omega_k^2$ depending on the isotope composition. One can show that for static ξ_e the relaxation rate is given by [59]

$$\tau^{-1}(\xi_e) = \frac{\Delta^2}{\xi_0} \, f(\xi_e/\xi_0) \, , \tag{4.4}$$

where $f(x)$ is a dimensionless function vanishing fast for large x. The nuclear spin diffusion mechanism provides a natural explanation to the effect of the isotope substitution on the hole width in the hole digging experiments [103, 7].

As mentioned above, the results of measurements at very high sweeping rates are universal: when the external bias is changing much faster than $\xi_N(t)$ the internal bias is simply absorbed to the time origin and we arrive at Eq. (4.3). However, when $\Delta^2 \ll \beta \ll d\xi_N/dt$, one may not exclude another plateau in $P\beta$. In this limit, the transition probability is small and one may consider only terms quadratic in Δ. Since the dynamics of the central spin is incoherent, all one has to do is to integrate the relaxation rate (4.4) over time (see also [60])

$$P \approx \int_{-\infty}^{\infty} \tau^{-1}(\beta t) \, dt = \pi C \Delta^2/\beta \, , \tag{4.5}$$

where $C = (1/\pi) \int_{-\infty}^{\infty} f(x) dx$.

An amazing result is that $C = 1$ for arbitrary stationary (time invariant) noise. In the path integral formulation of the ideal two-level dynamics the probability of finding the central spin state flipped at time t is given by

$$P(t) = \left| \sum_{n=0}^{\infty} (i\Delta)^{2n+1} \int_{t_0}^{t} \ldots \int_{t_{2n}}^{t} dt_1 \ldots dt_{2n+1} \, e^{-i\Phi} \right|^2 , \tag{4.6}$$

with the phase integral

$$\Phi = \int_{t_0}^{t} \xi_N(\tau)\eta(\tau)d\tau \tag{4.7}$$

$$\eta(\tau) = \begin{cases} 1 \text{ if } \tau \in (t_{2i}, t_{2i+1}) \\ -1 \text{ if } \tau \in (t_{2i-1}, t_{2i}) \text{ and } \tau \in (t_{2n}, t) \end{cases} . \tag{4.8}$$

In the LZ problem the time origin is set at the level crossing point $\xi_N(0) = 0$, $t_0 \to -\infty$, and $t \to \infty$. Now, the transition probability in second order in Δ is given by

$$\begin{aligned} P &= \Delta^2 \langle \left| \int_{-\infty}^{\infty} dt exp\left\{ -2i \int_0^t [\beta\tau + \xi_N(\tau)] d\tau \right\} \right|^2 \rangle \\ &= \Delta^2 \int\int_{-\infty}^{\infty} dt_1 dt_2 \, e^{i\beta(t_2^2 - t_1^2)} \, G(t_2 - t_1) \\ &= \Delta^2 \int_{-\infty}^{\infty} d\tau \, G(\tau) \int_{-\infty}^{\infty} dz \, e^{i2\beta z\tau} \equiv \frac{\pi G(0)\Delta^2}{\beta} , \end{aligned} \tag{4.9}$$

where G is the noise correlation function

$$G(t_2 - t_1) = \langle exp\left\{ i \int_{t_1}^{t_2} \xi_N(\tau) \, d\tau \right\} \rangle , \tag{4.10}$$

with an obvious property $G(0) = 1$.

The largest sweeping rate reported in [105] is about $\dot{B} = \beta/\mu_S \sim 1 \; T/s$. If one take $\xi_0/\mu_S \sim 8 \; mT$ for the limiting value of the hole width reported in Ref. [7], and the transverse nuclear spin relaxation time $T_2 \sim 1 \; ms$, then one will observe that experiments where most likely (it is hard to make a definite statement in the absence of data on the nuclear spin dynamics at low temperature) performed for $\beta \ll \xi_0/T_2$. In fact, the experimental plateau in $P\beta$ starts at much slower sweeping rates of order $\dot{B} > 0.01 \; T/s$. However, since the plateau value is not modified even in the fast noise regime, it is still possible to use an ideal expression for the data analysis. The down side is that spin diffusion alone may not explain the isotope effect.

B. Effect of the orthogonality blocking mechanism

Even if one ignores the last term in the Hamiltonian (spin diffusion) but considers a set of $\{\vec{\omega}_\perp^k\}$ with non-zero components in the directions orthogonal to $\hat{z}$, one still has a complex coupled dynamic system. As mentioned already, any change in the state of the central spin is then accompanied by the change in the direction of local field acting on $\{\hat{\vec{\sigma}}_k\}$, which, in turn, forces nuclear spins to precess, or, speaking quantum mechanically, to flip. In molecular magnets non-zero $\{\omega_\perp^k\}$ are mainly due to dipole fields of other molecules, and typically $\Omega_k \ll \omega_k$. We simplify the model further by considering identical and large hyperfine couplings $\omega_{||}^k = \omega_0 \gg \Delta$, then states of $2N$ nuclear spins may be classified in terms of polarization groups $M = \sum_k \sigma_z^k \in (-N, -N+1, \ldots, N)$, i.e.

$$\hat{H} = (\beta t + M\omega_0)\,\hat{\tau}_z + \Delta\,\hat{\tau}_x + \sum_k \hat{\sigma}_x^k \Omega_\perp^k\ . \tag{4.11}$$

We will concentrate on the dynamics of this model for sweeping rates $\beta \ll \omega_0^2$; otherwise the ideal answer (4.3) holds true. The advantage of the model (4.11) is that it allows an explicit analytic solution at each level crossing [58, 59] even when the spin bath polarization changes from M to M'. The spectrum of model (4.11) consists of $(2N+1)$ levels for each direction of the central spin, and the time dependence of energy levels is shown schematically in Fig.12. LZ transitions in this diagram happen at points $t \approx -(\omega_0/2\beta)(M+M')$, and correspond to resonances where S can either flip, i.e. switch to a level of the other band, or to stay on the same level. The corresponding dynamics is known as a motion on the "Landau-Zener grid".

For $\omega_0^2 \gg \beta$ we may isolate level crossings between states corresponding to the spin bath polarizations M and M'. Two approximations are involved in considering level crossings independently from each other. First, for multiple transitions at

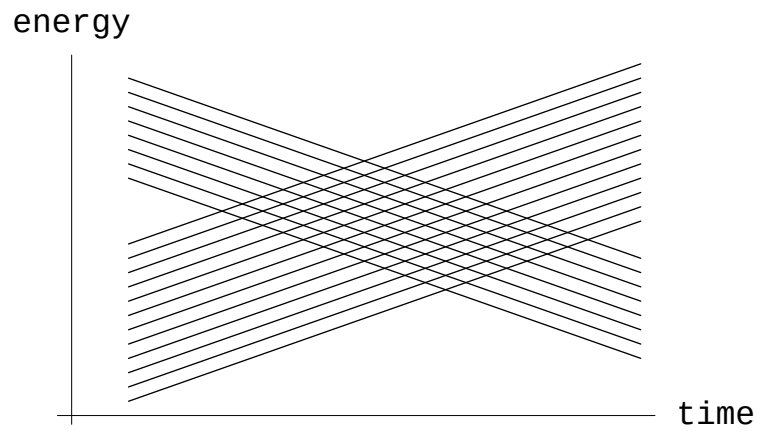

Fig. 12. Two intersecting energy bands representing different spin bath polarization groups.

different level crossings the time limits in integrals are restricted by the value of $\omega_0/\beta \gg 1/\omega_0$. Nevertheless, since the phase integral (4.8) remains large in this case, we may safely omit the above mentioned restrictions. Second, we ignore interference effects between level crossings. The dynamic coupling to the spin bath in the Hamiltonian (4.11) is characterized by $\kappa \approx \sum_k (\Omega_k/\omega_0)^2/2$ which may be interpreted as the average number of nuclear spins flipped when the central spin makes a transition [59]. If $\kappa = 0$, the matrix element to change the polarization state is zero and there is only one LZ transition at $M' = M$. If $\kappa \gg 1$, many environmental spins are flipped in each transition, and, excluding improbable cases when exactly the same nuclear spins are flipped again, the successive transitions do not interfere. Note, that *all* transitions with $M' \neq M$ necessarily involve flipping at least $|M' - M|$ environmental spins. Large phase integrals also work in favor of the second approximation.

For non-zero κ the path integral expression for $P_{M'M}$ has exactly the same form as Eq. (4.6), but now with Δ being replaced with [59]

$$\Delta^2 \to \Delta^2_{M'-M}(y) = \Delta^2 \, J^2_{M'-M}(2\sqrt{y\kappa}) \; , \tag{4.12}$$

where y is an integration variable such that the final answer has the form of a weighted integral over dy:

$$\begin{aligned} P_{M'M} &= \int_0^\infty dy\, e^{-y}\, P^{(0)}(\Delta_{M'-M}(y)) \\ &= \int_0^\infty dy\, e^{-y}\, \left(1 - e^{-\pi\Delta^2_{M'-M}(y)/\beta}\right) . \end{aligned} \tag{4.13}$$

Here $J_n(x)$ is the Bessel function of order n. In the regime $\Delta^2/\beta \ll 1$ all transition probabilities are small, and we may ignore multiple tunneling transitions. Then, the probability for the central spin to flip after all levels cross is given by the sum

$$P = \sum_{M'} P_{M'M} = \frac{\pi\Delta^2}{\beta} \int_0^\infty dy\, e^{-y} \sum_n J_n^2(2\sqrt{y\kappa}) = \frac{\pi\Delta^2}{\beta} , \tag{4.14}$$

which reproduces the ideal answer irrespective of the value of κ, though for $\kappa \gg 1$ most of the transitions contributing to (4.14) involve many nuclear spins flipped along with S. The origin of this result is in the unitary nature of matrix elements for the spin bath transitions [59].

Within the generic Hamiltonian (4.1), we may consider a model combining spin diffusion noise mechanism with the polarization group structure of the energy spectrum. Imagine that hyperfine frequencies have a distribution of values with a spread $\delta\omega_0 \ll \omega_0$ around the mean value ω_0, so that the notion of the polarization group M is still well defined, but the bias energy produced by this group has a noise component, $\xi_N(M,t) = M\omega_0 + \delta\xi_N(t)$, originating from flip-flip transitions in the spin bath. In the $\Delta^2/\beta \ll 1$ limit the solution of such a model is straightforward—at each level crossing tunneling is mediated by $\Delta^2_{M'-M}$ and the phase integral by $\delta\xi_N(t)$. The final result, obviously, reduces to Eq. (4.3) since the summation over n and the integration over time commute. The origin of the isotope effect in experiments [105] remains a puzzle because none of the spin bath mechanisms explains it.

We now switch to the study of the flipping probability at arbitrary Δ^2/β. At slow sweeping rate we must take into account multiple transitions between the bands of energy levels corresponding to different polarizations of the bath. For definiteness and simplicity, we consider the first band going "up" with the the lowest level having index $n = 1$ where $n = M + N + 1 = 1, 2, \ldots 2N + 1$ enumerates energy levels with different polarizations of the spin bath for the band going "up"; for the second band the highest level has index $m = 1$. LZ grids were considered previously in [4] with application for the Stark effect, but the evolution of the system on the grid rather than the total transition probability between two bands was investigated. As we show below, the latter calculation can be considerably simplified.

Since we are interested in the total probability of transfer to another band, the problem is more naturally formulated in terms of probabilities to remain on the same (first) band starting from the situation when the nearest crossing in time is between the levels n and m (we denote them $A_{n,m}$), and the complimentary probabilities B_{mn} to end up on the first band when starting on the level m of the second band and having level n for the nearest crossing. Then following recursion relations immediately derive from these definitions:

$$\begin{aligned} A_{n,m} &= p_{n,m} A_{n,m-1} + q_{n,m} B_{n-1,m} \\ B_{n,m} &= \bar{p}_{n,m} B_{n-1,m} + \bar{q}_{n,m} A_{n,m-1} \end{aligned} \tag{4.15}$$

where $q_{n,m} = 1 - p_{n,m}$, $\bar{q}_{n,m} = 1 - \bar{p}_{n,m}$, and p and $\bar{p}$ are the probabilities to stay on the same level for the first and second bands. Recursion relations (4.15) together with conditions $B_{0,m} = 0$, $A_{n,0} = 1$ and transition probabilities (4.13) provide a tool for a simple numerical solution of the problem.

For the spin bath temperature much higher than hyperfine couplings the proba-

bilities p and $\bar{p}$ coincide. Furthermore, for the model (4.11) the coefficients $q_{n,m}$ can be taken from the formula (4.13). In our numerical calculations the bath was supposed to be unpolarized in the initial state, $n = N + 1$, i.e. the number of nuclear spins along and opposite to the central spin were equal, and the nearest crossing index was $m = 2N + 1$. In Fig.13 we show the probabilities to keep the same direction of S for different values of κ obtained by numerically solving equation (4.15) for $N = 150$. Larger values of N or non-zero but small initial polarization of the spin bath do not change the results. At small values of Δ^2/β, i.e. for large sweeping rates, the transition probabilities converge to the ideal LZ formula. For $\Delta^2/\beta > 0.5$ the effect of the spin bath is noticeable even for rather small values of $\kappa > 0.1$. The results saturate for $\kappa > 1$, and curves with $\kappa = 1$ and $\kappa = 15$ are hardly distinguishable from each other.

Though all transitions are incoherent, the probability for S to flip can be larger than 1/2 even in the strong coupling regime $\kappa >> 1$. This is because initially the system was in the middle of the fist band. There is a finite probability to tunnel to any level of another band but transitions to levels of the same band having higher energies than the initial one are forbidden. Hence there are more states to tunnel to another band. This result is distinct from predictions of other approaches to incoherent Landau-Zener theory. For example, in the limit of strong decoherence due to the coupling to strong diagonal noise the transition probability approaches 1/2 at large Δ [50].

At large values of κ and $\beta \sim \Delta^2$ we enter an interesting regime when transition probabilities at one resonance are small but the number of resonances with non-vanishing transition probabilities is large. For small $q_{n,m}$ and large number of states on the grid we can switch to continuous variables $n \to y$, $m \to x$ and introduce the transition probability density $\alpha(x, y)$. If the occupied level y passes a region of a small

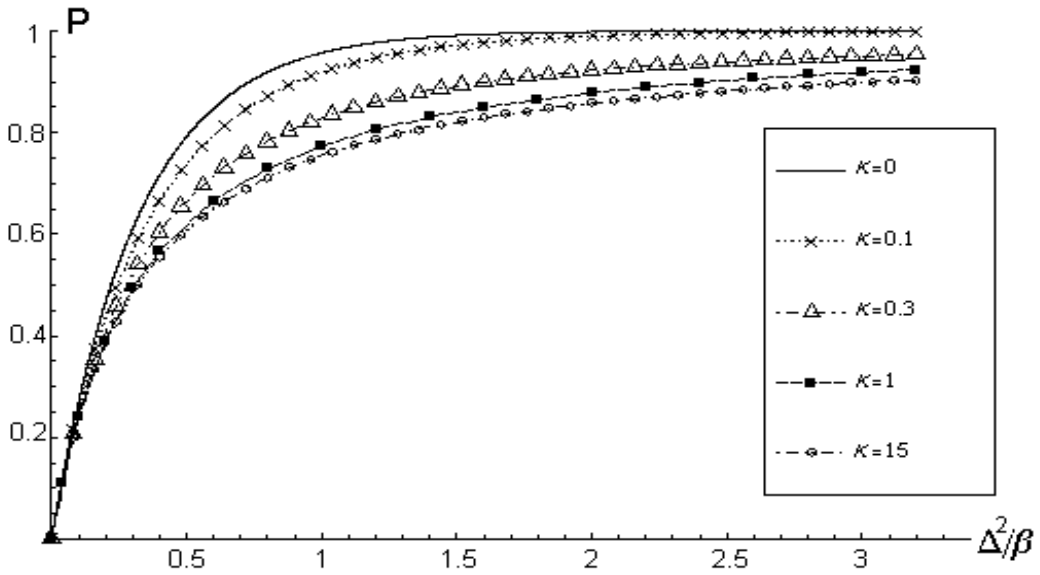

Fig. 13. The probability for the central spin to flip in the model of incoherent Landau-Zener transitions as a function of Δ^2/β. The solid line represents the "ideal" curve $P = 1 - e^{-\pi\Delta^2/\beta}$.

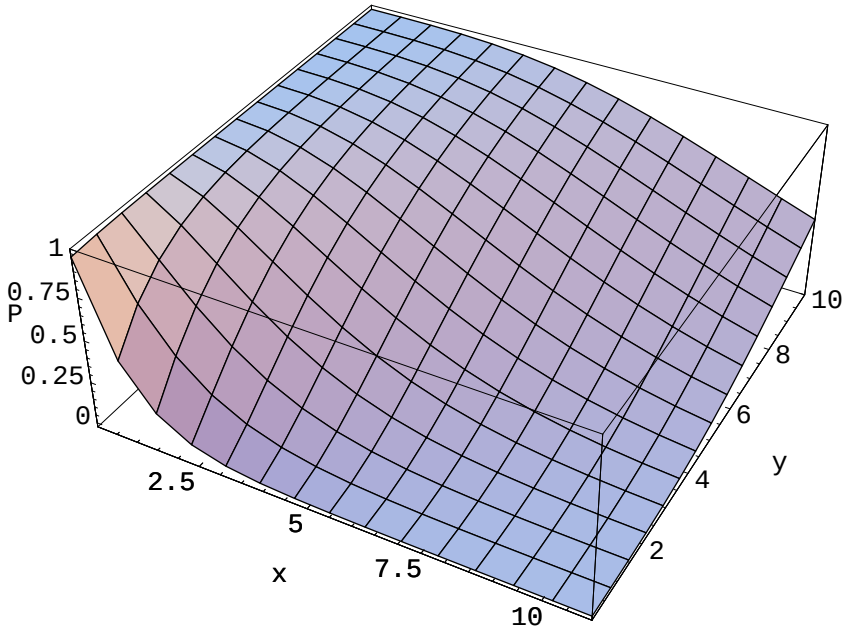

Fig. 14. The probability to stay on the same band in the continuous limit as a function of the band thickness. The energy scale is normalized to $1/\alpha$.

thickness dx of the second band, the probability to flip S is given by $\alpha(x,y)dx$. Let $A(x,y)$ is the probability to end up on the same (first) band if the system is initially on the first band at point (x,y). Respectively, $B(x,y)$ is the probability to endup on the first band when being initially on the second band at point (x,y). Finally, the probability to stay on the same level during the whole process is $e^{-\int_0^x \alpha(x,y)dx}$. It is convenient to introduce functions $f(x,y)=\int_0^x \alpha(t,y)dt,\ g(x,y)=\int_0^y \alpha(x,t)dt$. Then the integral equation for transition probabilities between the two bands reads:

$$\begin{aligned} A(x,y) &= e^{-f(x,y)} + \int_0^x e^{-(f(x,y)-f(u,y))}\alpha(u,y)B(u,y)du \\ B(x,y) &= \int_0^y e^{-(g(x,y)-g(x,v))}\alpha(x,v)A(x,v)dv \end{aligned} \tag{4.16}$$

this equation is equipped with boundary conditions $A(0,y)=1,\ B(x,0)=0$.

To have a deeper insight into final answers, we consider an explicit solution for a

simplified model that is described by equation (4.16) with constant $\alpha(x, y) = \alpha$. The solution can be written in a closed form:

$$P(x, y) = e^{-\alpha x}(1 + \alpha\sqrt{x}\int_0^y e^{-\alpha t}\frac{I_1(2\alpha\sqrt{xt})}{\sqrt{t}}dt) \,, \tag{4.17}$$

where $I_1(x)$ is the modified Bessel function. We visualize it in Fig.14. The plot demonstrates that if the number of states to cross in the second band is larger than in the first band, i.e. $y < x$, the probability to stay on the same band can be made arbitrarily small by varying the sweeping rate that controls the value of αx.

C. Comparison with numerical simulations

Quantum dynamics of a central spin interacting with the spin bath is very complex. To make it analytically tractable, many works replace the spin bath by a randomly varying magnetic field acting on the collective electronic spin of a magnetic molecule. However, its use may lead to considerable errors [115]. To avoid any kind of noise approximations we directly simulated dynamics of both the central spin and all the spins of the bath by solving the time-dependent Schrödinger equation for the whole system [116, 117, 118]. Using this approach, we addressed validity of the noise approximation, and showed that in many cases it is not valid.

We have used two methods suitable for studying many-spin systems, one employing the Chebyshev polynomial expansion [116, 117], and the other based on the Suzuki-Trotter formula [118]. We chose the following hierarchy of parameters which qualitatively reproduces the realistic magnetic molecules. The dipolar interactions between the nuclear spins were assumed to be uniformly distributed in the interval $0 < V_{ij}^{\alpha\beta} < 0.5$. The smallest energy parameter is the tunneling amplitude; it was set $\Delta = 0.25$. The strength of the hyperfine interactions is much stronger, $\omega_{||}^{i} \gg \Delta, V_{ij}^{\alpha\beta}$,

so we took $\omega_{||}^i$ uniformly randomly distributed over the interval from 0 to 20, i.e. the average value of this parameter is 10. The orthogonality blocking parameters $\omega_{\perp}^i$ are chosen comparable with the hyperfine couplings $\omega_{||}^i$, to represent the case when the parameter κ is not small (as in realistic magnetic molecules), and where analytical treatment is complicated. Simulations were performed for $N = 12$ and sometimes for $N = 16$ nuclear spins 1/2 interacting with the central electronic spin represented as a pseudo-spin 1/2 system. In the simulations we assume that the vector $\vec{\omega}_{\perp}$ is directed along the x axis. It can also have nonzero y and z projections, but our simulations show that this does not lead to qualitative changes.

During the field sweep, the system initially being in the state $|\downarrow\rangle$ has a finite probability to occur in the state $|\uparrow\rangle$. For an isolated two-level system, this probability is the Landau-Zener probability but for a system interacting with its environment, the transition probability can deviate from this result. In previous subsection, to make the problem of spin bath dynamics analytically tractable, we invoked a number of approximations (all $\omega_{||}^i$ were assumed equal, the noise and the orthogonality blocking mechanism were considered separately). Hence it is important to investigate the Landau-Zener transition probability without such approximations.

If the influence of the system's environment can be represented as a strong and fast diagonal noise, the standard LZ prediction (4.2) is no longer valid; it has been shown by Kayanuma [46] that in that case the transition probability is

$$P_K = \frac{1 - \exp\left(-2\pi\Delta^2/\beta\right)}{2} \tag{4.18}$$

This formula is valid for example, when the two level system at a Landau-Zener intersection is coupled to phonon bath at high temperature [50]. However, according to [61], for a system strongly coupled to a nuclear spin bath, equation (4.18) does not necessarily describe the LZ transitions even in the presence of strong decoherence.

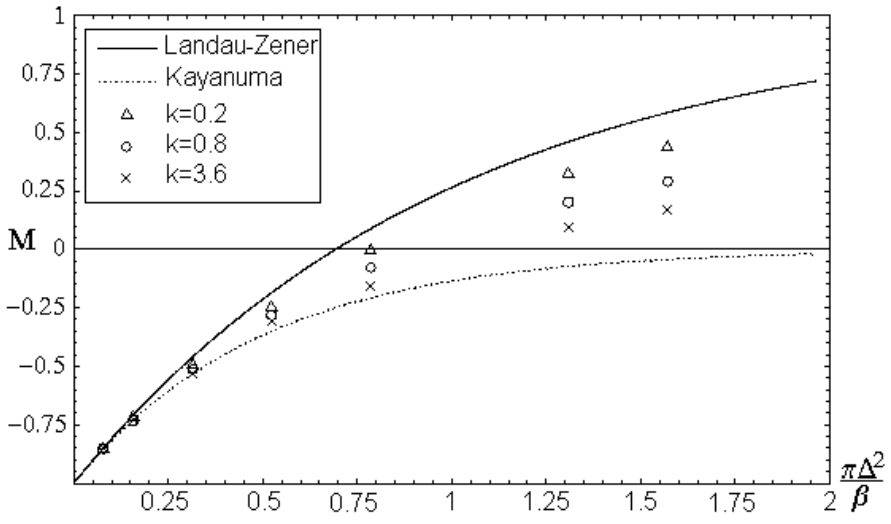

Fig. 15. Magnetization of the central spin after a linear sweep of magnetic field for different strengths of orthogonality blocking mechanism and in the absence of the nuclear spin diffusion. The initial magnetization is normalized to -1.

Rather a description in terms of incoherent transitions on the LZ grid [4] can be more relevant. In Fig.15 we present the results of our simulations for different values of the orthogonality blocking parameter κ. The transition probability P is directly related to the final magnetization of the central spin $M = \langle \tau_z \rangle$ after the sweep, $M = 2P - 1$. The results in Fig.15 are in good agreement with the conclusions of [61], even though our simulations do not assume equal strengths of the hyperfine couplings. At fast sweeping rates ($\Delta^2/\beta \ll 1$), the standard LZ prediction is correct, but at slower sweeps the final magnetization deviates toward the prediction of Eq. 4.18.

The simulations show, the final magnetization can be positive for $\kappa > 1$, which means that the transition probability is larger than 1/2.

As it is explained in previous subsection, this result can be interpreted in terms of the multiple level crossings. In the course of the LZ process, due to interaction with the spin bath, the two central spin levels are split into two crossing bands,

where every energy level of the band represents some state of the nuclear spin bath. The system starts in some state of the band going up in energy. During the field sweep, it is forbidden for the system to make the transitions to those states of the same band which have energies larger than the energy of the initial diabatic state. However, there is no such restriction for the transitions to states of another band. Therefore, statistically, there are more possibilities for the system to change the band after passing through all intersections of the levels in the bands.

If properly rescaled, the results in Fig.15 would be in a very good agreement with analytical predictions in Fig.14. Therefore we conclude that the description of the coupling to the spin bath at level intersection in terms of incoherent transitions on a Landau-Zener greed very well catches the related physics.

CHAPTER V

SUMMARY AND CONCLUSIONS

1. The time-dependent behavior of nanoscale systems usually cannot be treated in approximations of weak perturbations. The Landau-Zener theory is one of the most powerful tools in nonstationary quantum mechanics that allows investigation analytically of strongly driven systems with explicit time-dependence of the parameters. However conventional Landau-Zener theory was designed to deal with a small number of quantum states. In nanoscale systems the number of degrees of freedom that must be considered is not macroscopic, but it usually cannot be considered as small. Also, in reality the coupling of such systems to degrees of freedom of the environment cannot be disregarded. This forced us to search for useful generalizations of the Landau-Zener theory.

2. One of the possibilities is to generalize the two-state Landau-Zener formula to the case of multistate evolution. However the solution of the multistate Landau-Zener model is unknown except for a few exactly solvable classes, some of which are too artificial to have realistic physical interpretations. During our study we found new classes of exactly solvable multistate Landau-Zener models, all of which have possible realistic applications. The first such solvable class has the interpretation of the transitions of electrons in a linear chain of sites, driven by an external homogeneous time-dependent electric field. It was solved by employing symmetry of the Hamultonian under translation which allowed reduction of the infinite chain of equations for amplitudes to a first order partial differential equations. The second achievement is the conjecture of the absence of counterintuitive transitions in any model with the Hamiltonian

(2.1). Besides demonstrating numerical tests that confirmed it, we showed that this conjecture, together with the Brundobler-Elser hypothesis, can be proved by analytical continuation of the evolution into complex times. The absence of counterintuitive transitions is important to explain the peculiarities of the Landau-Zener transitions in a two level system coupled to the spin bath or the oscillator bath at low temperature. The third finding is the symmetry relating many solvable multistate models that allows generation of new exactly solvable Landau-Zener models from already known ones by considering the evolution of operators rather than of amplitudes and then projecting it onto the Hilbert space.

We demonstrated possible applications of the generalizations of the Landau-Zener evolution to the multiparticle sector. One of the solvable fermionic models describes the charge transfer through the energy level of a quantum dot. The bosonic models can be employed in the physics of Bose condensates. For example, we considered a time dependent problem of coherent dissociation of the molecular condensate into distinct modes near the Feshbach resonance working beyond the mean field approximation. Our approach allowed investigation of this process nonperturbatively.

Despite the fact that the multistate Landau-Zener problem remains generally unsolved, one can make general conclusions on the basis of the recent results. On the one hand some elements of the scattering matrix are known in any multistate model. This justifies speculations about existence of the complete solution of the problem. Such a solution, if found, would be one of the most important results in nonstationary quantum mechanics. On the other hand, all exact solutions found have much in common, namely that they can be gen-

erated by the simple empirical rule according to which one should apply the two state Landau-Zener formula at every pair of level intersections. Hence it is possible that instead of the complete solution there is a special class of solvable models whose transition probabilities can be found by successive application of the two-level formula. Then there might be a symmetry in the background of all such solvable models which allows reduction of their solution to a genuine Landau-Zener result. Understanding of this symmetry may also have interesting consequences in mathematical physics. In any case more mathematical study is needed and interesting results are anticipated.

3. In order to model the coupling of the Landau-Zener system with the environment we added an additional parameter to the Landau-Zener Hamiltonian that describes interaction of the central two level system with classical stationary noise. In quantum optics and atomic physics the traditional approach to a system coupled to external degrees of freedom is to write an effective master equation with phenomenological parameters that are responsible for decoherence or spontaneous transitions. Such a simple approach in most cases fails to predict correctly the Landau-Zener transition probabilities because the distance between diabatic states linearly increases with time and the effect of noise depends on this energy distance. In other words, one cannot assume that the phenomenological parameters in the master equation are time-independent. We designed the averaging technique that allowed us to find the true master equation and its simple analytical solutions for the most general case and for all possible nontrivial limits. Also, we designed a theoretical technique, based on the Bloch tensor formalism which led to the generalization of our results to the Landau-Zener transitions of arbitrary spin in regular and noisy fields. The

validity of our results are confirmed by numerical simulations.

4. In molecular nanomagnets the tiny tunneling amplitude can be derived from the measurements of the Landau-Zener transition probability in the sweeping magnetic field. Even at low temperatures molecular nanomagnets are relatively strongly coupled to the nuclear spins which results in quick decoherence and other effects related to nuclear spin assisted tunneling. Measurements demonstrated the isotope effect, namely dependence of the transition probability on the average number of nuclear spins in the molecules. However, the isotope effect is not so strong as to destroy such coherent phenomena as oscillation of the tunneling amplitude as function of external transverse magnetic field. To find the explanation of the experimental results, we considered the Hamiltonian of a central two level system coupled to the bath of nuclear spins. Interaction of nuclear spins with each other results in fluctuations of the effective hyperfine field acting on the central system. This effect can be described as a stationary noise acting on the two-level system. Another effect is due to the possibility of the nuclear spin co-flips due to strong interaction of nuclear spins with the central system. According to results of our analytical and numerical study none of the decoherence mechanisms can explain the isotopic effect observed at fast sweeping rates of the external field, i.e. the Landau-Zener formula is robust at fast sweeps, even when coupling to the spin bath is relatively strong. This fundamental contradiction between the theory and the experiment demonstrates that the dynamics of the central spin coupled to a spin bath is still poorly understood. At slower sweeping rates we predict strong deviations from the Landau-Zener formula. The transition probability depends on which decoherence mechanism dominates. If the effect of spin bath dynamics is equivalent

to a strong stationary noise the transition probability cannot be larger than 1/2. However, in most studied magnetic molecules the central spin is strongly coupled to the bath so that the bath dynamics is slaved to the dynamics of the molecular spin. The strong decoherence then is due to the nuclear spin flips allowed during the tunneling in the central system. We proposed that such a tunneling can be adequately described by the incoherent the Landau-Zener transitions on the greed of intersecting levels, where each level corresponds to a distinct state of the spin bath and belongs to one of the two bands that appear due to splitting of two energy levels of the central system. The numerical simulations confirmed the analytical predictions of this model. The main feature of domination of the orthogonality blocking mechanism is the possibility of a transition probability larger than 1/2 even in the limit of strong decoherence and slow sweeping rate of the external field.

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