VIBRATIONAL SCHROEDINGER CATS

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Abstract

The optical Schroedinger cat states are simple realizations of quantum states having non-classical features. It is shown that vibrational analogues of such states can be realized in an experiment of double pulse excitation of vibrionic transitions. To track the evolution of the vibrational wave packet we derive a non-unitary time evolution operator so that calculations are made in a quasi Heisenberg picture.

1   Introduction

The analog of the classical harmonic oscillation in the quantum mechanics is the coherent state $| \alpha \rangle$ defined as an eigenstate of the annihilation operator $b | \alpha \rangle = \alpha | \alpha \rangle$. Both in the position and in the momentum representations the absolute square of its wave function has a Gaussian shape. It performs harmonic vibration in time with an amplitude that depends on the initial excitation. The superposition of two coherent states [1]

$$| + \rangle = N_{|+\rangle}(| \alpha \rangle + | -\alpha \rangle),$$ (1)

$$N_{|+\rangle} = \frac{1}{\sqrt{2 + 2e^{-2\alpha^2}}},$$

situated sufficiently far from each other in the phase-space can be considered as the superposition of two macroscopically distinguishable quasiclassical states called Schroedinger cat state.

Recently great interest has been paid to such superposition states in quantum optics [2-11]. Non-classical features of Schroedinger cat states i.e. squeezing, [4] sub-Poissonian statistics, oscillation in photon statistics, etc. were discussed rather widely. It was shown [5, 7] how the quantum interference between the coherent states involved in the superposition leads to the occurrence of non-classical features. Due to the interference a fringe pattern appears between the Gaussian bells representing the coherent states in the Wigner function picture. This fringe pattern is transformed characteristically when the positions or the number of the coherent states changes. There are several promising schemes to produce nonclassical states of light using the concept of Schroedinger cat states [8, 9].

A wide interest was addressed to wave packet formation and motion during Franck-Condon transitions in both theoretical and experimental points of view [4, 10, 11]. In this paper we shall discuss the possibilities of producing Schroedinger cat like superpositions of the vibrational states during Franck-Condon vibrionic transitions in molecules or in crystals. As we shall see such states can be created by two short pulses separated in time appropriately.

2   The model Hamiltonian

Let us consider a one-vibrational-mode model specified by the adiabatic Hamiltonians

$$\hat{H}_i = \epsilon_i + \frac{\hat{p}^2}{2M} + \frac{M\omega_i^2}{2}(\hat{q} + q_i)^2, \tag{2}$$

$$\hat{H}_e = \epsilon_e + \frac{\hat{p}^2}{2M} + \frac{M\omega_i^2}{2}\hat{q}^2, \tag{3}$$

corresponding to the molecular vibrations in initial (i) and excited (e) electronic states. Here $\epsilon_{i,e}$ are electronic energy levels and $\omega_{i,e}$ vibrational frequencies.

In terms of the annihilation phonon operators b associated with the vibrational potential of the excited states,

$$\hat{q} = \sqrt{\frac{\hbar}{2M\omega_e}}(\hat{b}^\dagger + \hat{b}), \quad \hat{p} = i\sqrt{\frac{\hbar M\omega_e}{2}}(\hat{b}^\dagger - \hat{b}), \tag{4}$$

the Hamiltonians of Eq. (2,3) have the forms

$$\hat{H}_i = \epsilon_i + \frac{M\omega_i^2}{2}q_i^2 + \hbar\omega_i\{\frac{1}{4}(\frac{\omega_i}{\omega_e} + \frac{\omega_e}{\omega_i})(\hat{b}^\dagger\hat{b} + \hat{b}\hat{b}^\dagger)+$$

$$+\frac{1}{4}(\frac{\omega_i}{\omega_e} - \frac{\omega_e}{\omega_i})(\hat{b}^\dagger\hat{b}^\dagger + \hat{b}\hat{b})\} + \frac{\hbar\omega_i^2 q_i}{\omega_e}\sqrt{\frac{M\omega_e}{2\hbar}}(\hat{b}^\dagger + \hat{b}) \tag{5}$$

$$\hat{H}_e = \epsilon_e + \frac{1}{2}\hbar\omega_e(\hat{b}^\dagger\hat{b} + \hat{b}\hat{b}^\dagger). \tag{6}$$

The Hamiltonian of the initial state can be diagonalized by the unitary operator

$$\hat{\Sigma} = e^{-g(\hat{b}^\dagger - \hat{b})}e^{r[\hat{b}^2 - \hat{b}^{\dagger 2}]/2}, \tag{7}$$

$$g = q_i\sqrt{\frac{M\omega_e}{2\hbar}}, \quad r = \frac{1}{2}ln\frac{\omega_i}{\omega_e}.$$

Here g and r are displacement and squeezing parameters correspondingly. The vibrational ground state of the initial electronic level is

$$| 0\rangle_i = \hat{\Sigma} | 0\rangle_e, \tag{8}$$

where $| 0\rangle_e$ is the vibrational ground state of the excited electronic level.

The Hamiltonian $\hat{H}'(t)$ describing the interaction with the external field has the form

$$\hat{H}'(t) = \frac{1}{2}d_{ie}E(t)\hat{a}_e^\dagger\hat{a}_i + \frac{1}{2}d_{ie}^*E^*(t)\hat{a}_i^\dagger\hat{a}_e, \tag{9}$$

where

$$E(t) = e(t)exp(-i\Omega_0 t),\tag{10}$$

$\hat{a}_{i(e)}$ is the annihilation operator of the $i(e)$-th electron level, d_{ie} the dipole matrix element of the electronic transition, $\mid e(t)\mid^2$ and Ω_0 are the envelope function and the central frequency of the exciting pulse.

Suppose the electronic transition takes place instantaneously. The emerging vibrational wave packet is described by Eq. (8). The time evolution of the wave packet in the excited level is driven by the unitary operator $\exp[-(i/\hbar)\hat{H}_e t]$. So the evolution of the vibration from $-t_0$ until t is described by the unitary operator

$$\hat{G}(t - t_0) = e^{-\frac{i}{\hbar}\hat{H}_e(t-t_0)}\hat{\Sigma}.\tag{11}$$

Let us assume that initially, at $t = -\infty$ the system is in the ground state $\mid i\rangle \mid 0\rangle_i$. After the exciting pulse has passed according to the first order perturbation theory the electronic-vibrational wave function takes the form:

$$\mid \Psi, t\rangle = \mid i, t\rangle \mid 0\rangle_i - i\frac{E_0 d_{ie}}{2\hbar}\mid e, t\rangle \mid \{E(t)\}\rangle_e.\tag{12}$$

Here $\mid \{E(t)\}\rangle_e$ is unnormalized vibrational wave function of the molecule in the excited electronic state:

$$\mid \{E(t)\}\rangle_e = \int_{-\infty}^{t} d\tau E(\tau)e^{i\Delta\tau}\hat{G}(t - \tau) \mid 0\rangle_e,\tag{13}$$

where $\Delta = \bar{\Omega} + (\omega_e - \omega_i)/2$, $\bar{\Omega} = (\epsilon_e - \epsilon_i)/\hbar$. The timedependent part of $\hat{G}(t)$ is the exponential of $\hat{H}_e$. Assume the pulse duration is short compared with the observation time t. In this case we can put the upper limit of the integration to infinity. This condition means we perform measurements after the excitation pulse has passed. The integration in Eq. (13) can be done explicitly. Separating the operator in Eq. (13) in front of the vacuum the non-unitary time evolution operator is

$$\hat{T}(t) = \exp\left(-\frac{i}{\hbar}\hat{H}_e t\right)\exp\left[-\frac{(\hat{H}_e/\hbar - \delta)^2}{2u^2}\right]\hat{\Sigma},\tag{14}$$

where $\delta = \Delta - \Omega_0$.

In the following sections we shall investigate the properties of the vibrational wave function of Eq. (13) considering twin exciting laser pulses. For the sake of simplicity we suppose that there is no change of the vibrational frequency due to the electronic transition ($\omega_e = \omega_i = \omega$). In this case the operator $\hat{\Sigma}$ in Eq. (13) simplifies to a displacement operator $\hat{D}$ and the excited vibrational wave function has the form

$$\mid \{E(t)\}\rangle_e = \exp\left(-\frac{i}{\hbar}\omega_e\hat{n}t\right)\exp\left[-\frac{(\omega_e\hat{n} - \delta)^2}{2u^2}\right]\mid g\rangle_{coh},\tag{15}$$

where $\mid g\rangle_{coh}$ is a coherent state with respect to the phonon operator $\hat{b}$.

3 Double pulse excitation

Let us consider two identical Gaussian shaped pulses following each other by an interval T_1

$$E(t) = \frac{E_0}{2\pi^{\frac{1}{4}}} e^{-\frac{u^2}{2}(t+\frac{T_1}{2})^2 - i\Omega_0(t+\frac{T_1}{2})} + \frac{E_0}{2\pi^{\frac{1}{4}}} e^{-\frac{u^2}{2}(t-\frac{T_1}{2})^2 - i\Omega_0(t-\frac{T_1}{2}) - i\phi}, \tag{17}$$

here ϕ is a possible additional phase difference between the subpulses.

The vibrational state produced by such a twin pulse excitation has the form

$$\mid \{u, T_1, \delta, \phi\}\rangle_e = e^{-i\Omega_0 \frac{T_1}{2}} \mid u, (t + \frac{T_1}{2})\rangle + e^{i\Omega_0 \frac{T_1}{2} - i\phi} \mid u, (t - \frac{T_1}{2})\rangle, \tag{18}$$

$$\mid u, t\rangle = \exp\left(-\frac{i}{\hbar}\omega_e \hat{n} t\right) \exp\left[-\frac{(\omega_e \hat{n} - \delta)^2}{2u^2}\right] \mid g\rangle_{coh} \tag{19}$$

To investigate the quantum properties of the superposition state of Eq. (18) it is convenient to consider its Wigner function

$$W(\alpha) = \frac{1}{\pi^2} \int e^{\eta^* \alpha - \eta \alpha^* - \frac{1}{2}|\eta|^2} {}_e\langle\{u, T, \phi\} \mid e^{\eta b^\dagger} e^{-\eta^* b} \mid \{u, T, \phi\}\rangle_e \, d^2\eta. \tag{20}$$

For extremely short pulses we have coherent superposition states which are the vibrational analog of the so called optical Schroedinger cat states. The Wigner function and the time dependence of the absolute square of the wave function are shown in Fig. 1a and Fig. 1b correspondingly. The Wigner function consists of two bells of the superposed coherent states and an interference fringe between them. If the coherent states are far away the fringe has a lot of well-pronounced peaks. On the contrary, if the coherent states are near enough the fringe has only few peaks. In this case the fringe can partially merge with the bells and, depending on the phase between the component states, may decrease the uncertainty of one of the quadratures $\hat{X}_+ = b + b^\dagger$ or $\hat{X}_- = -i(b - b^\dagger)$ below the vacuum level.

4 Discussion

Baumert *et. al.* first excited the Na_2 molecule by a short laser pulse [12]. Applying a second laser pulse they excited the state once more. Depending on the time delay between the two successive pulses they had a molecule on another excited level or dissociated fragments. We suggest a similar experiment with a double pulse primary excitation leading to a Schroedinger vibrational state on the level e (Fig. 2). Applying a third pulse when the two parts of the Schroedinger cat state are furthest from each other one obtains a superposition of a molecule with its fragment.

This chemical cat state can lead us very near to the original paradox of Schroedinger. Let us suppose that this molecular superposition is superposition of the undamaged form of a virus's DNA with a denaturalized variant of the same virus. The resulting 'Schroedinger virus state' would be, in fact, a quantum mechanical superposition of a "living" and a 'dead' virus.
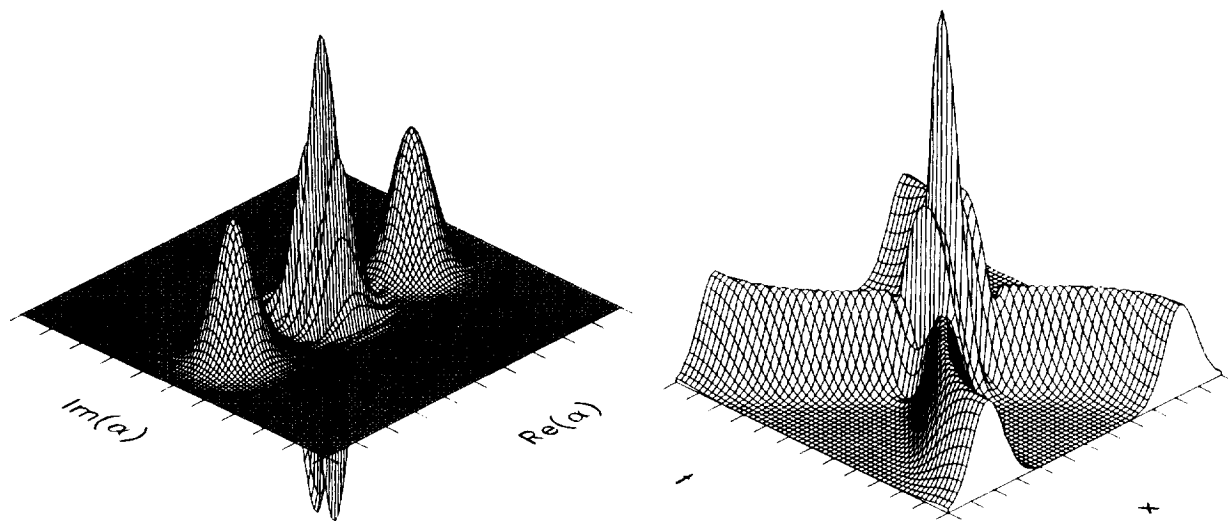
Acknowledgments

Fig. 1 The Wigner function (Fig. 1a) and the time-dependence of the absolute square of the wave function $| \psi(q) |^2$ (Fig. 1b) of the Schroedinger cats state. The prominent fringe structure between the coherent states' Gaussian bells of the Wigner function is caused by the quantum interference between the two parts of the superposition state. A similar interference fringe of the wave function can be found around $t = \pm \frac{T}{4}$, otherwise in the bigger part of the period $| \psi(q) |^2$ consists of two Gaussians representing the two superposed coherent states.
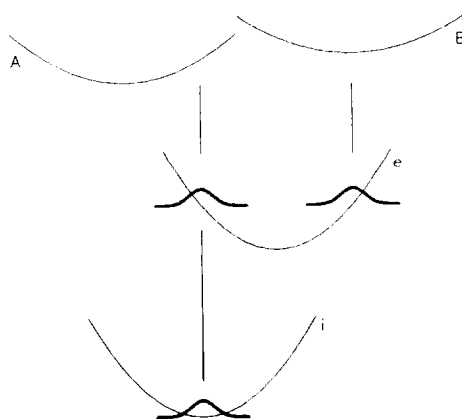

Fig. 2 The schematic terms for creation of a chemical superposition state. First either by double or appropriately chirped single pulse one prepares a vibrational superposition state on level e. At some moment of its separation by some secondary pulse(s) one can transfer the molecule into molecule A represented by the upper left term and simultaneously into molecule B shown as the upper right term, creating this way a chemical "Schroedinger cat" state.

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