

THE INTERFERENCE OF THE DYNAMICALLY SQUEEZED VIBRATIONAL WAVE PACKETS

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An electronic excitation of a molecule by a sequence of two femtosecond phase-locked laser pulses is considered. In this case the interference between the vibrational wave packets induced by each of the subpulses within a single molecule takes place. It is shown that due to the dynamical squeezing effect of a molecular vibrational state the interference of the vibrational wave packets allows one to measure the duration of a femtosecond laser pulse. This can be achieved experimentally by measuring the dependence of the integral fluorescence of the excited molecule on the delay time between the subpulses. The interference can lead to a sharp peak (or to a down-fall) in that dependence, the width of which is equal to the duration of the laser pulse. It is shown that finite temperature of the medium is favorable for such an experiment.

Recently a great interest has been shown to the study of spatially localized vibrational wave packets in molecules induced by ultrashort laser pulses. The time evolution of the mean position of such a wave packet corresponds to the nearly classical nuclei motion, and can be observed in the pump and probe spectroscopic optical experiments [1-4]. The idea of controlling the chemical reactions due to the possibility of the nearly classical nuclei motion has been widely discussed and experimentally verified [5-10].

An important characteristic feature of the vibrational wave packet is its spatial extent. In the case of a harmonic nuclear potential the spatial properties of the wave packet are closely connected with the phonon statistics and can be treated by the methods developed in quantum optics. In a series of previous publications [11-16] we have studied the statistical properties of the vibrational states appearing due to the excitation of Franck-Condon transitions by the transform-limited light pulses of finite duration. The problem was to learn how is it possible, by varying the characteristics of a laser pulse (i.e. its amplitude and phase modulation), to excite the molecular vibrations with the given quantum fluctuations of the conjugated variables. In particular, it was theoretically predicted [11-13] and experimentally verified [17] that, when the spectral width of the exciting pulse is smaller than that of the absorption band, there appears a molecule in a squeezed vibrational state with a reduced quantum uncertainty of the nuclei position. The mechanism of squeezing, which arises here, is of the dynamical nature and can be explained as the result of quantum interference in the phase space of the molecular vibrations [13]. An application of the dynamical squeezing effect to the problem of wave packets optimal shaping for the control of the chemical reactions was considered in [18-21].

Our goal now is to show how this dynamical squeezing effect can be put to use in another way for the duration measurement of a femtosecond laser pulse. Following [22], we shall consider the

excitation of a Franck-Condon transition in a molecule by a sequence of two femtosecond phase-locked Gaussian-shaped pulses:

$$\mathbf{E}(t) = \frac{\mathbf{E}_0}{2\pi^{1/4}} \left\{ \exp\left[-\frac{u^2}{2}\left(t + T/2\right)^2\right] + \alpha \exp\left[-\frac{u^2}{2}\left(t - T/2\right)^2\right] \right\} \exp(-i\Omega t) + c.c., \quad (1)$$

separated from each other by the time interval T . Here α is a possible additional complex coefficient between the pulses: $\alpha = |\alpha| \exp(i\varphi)$. As it was demonstrated experimentally in [22] it is possible to change the time interval T , keeping constant the value of phase φ .

We shall assume that due to the electronic transition only a spatial shift of the harmonic nuclear potentials occurs in the molecule. The adiabatic Hamiltonians describing molecular vibrations in the initial (i) and excited (e) electronic states have the form

$$H_i = \varepsilon_i + \frac{\hat{p}^2}{2m} + \frac{m\omega^2}{2}(\hat{q} + q_i)^2, \quad (2)$$

$$H_e = \varepsilon_e + \frac{\hat{p}^2}{2m} + \frac{m\omega^2}{2}\hat{q}^2, \quad (3)$$

where $\varepsilon_{i,e}$ are the electronic energy levels. It is convenient to rewrite Eqs. (2) and (3) expressing \hat{q} and \hat{p} through the phonon creation and annihilation operators:

$$\hat{q} = \sqrt{\frac{\hbar}{2m\omega}}(b^+ + b), \quad \hat{p} = i\sqrt{\frac{\hbar m\omega}{2}}(b^+ - b). \quad (4)$$

Then

$$H_i = \varepsilon_i + \frac{m\omega_i^2 q_i}{2} + \frac{\hbar\omega}{2}(b^+ b + b b^+) + \hbar\omega g(b^+ + b), \quad g = q_i \sqrt{\frac{m\omega}{2\hbar}}, \quad (5)$$

$$H_e = \varepsilon_e + \frac{\hbar\omega}{2}(b^+ b + b b^+). \quad (6)$$

A non-dimensional coupling constant g , which appears in Eq. (4), is equal to the ratio of the Franck-Condon shift to the amplitude of zero vibrations. The Hamiltonian (5) can be diagonalized by means of the unitary displacement operator

$$D = \exp[-g(b^+ - b)], \quad (7)$$

$$D^+ H_i D = \varepsilon_i + \frac{\hbar\omega}{2}(b^+ b + b b^+). \quad (8)$$

It is easily seen from Eq. (8) that the ground state vibrational wave function of the molecule in the initial electronic state has the form

$$|\Phi_i\rangle = D|0\rangle, \quad (9)$$

where $|0\rangle$ is the phonon vacuum, which coincides with the ground vibration state of the electronically excited molecule. Let us note that the displacement operator acting on the vacuum state gives the coherent state. Then Eq. (9) simply means that the ground state wave function of the harmonic oscillator being placed in the shifted potential is a coherent state.

The case of zero temperature. We shall assume that initially at $t = -\infty$ the molecule was prepared in the state $|i\rangle|\Phi_i\rangle$, where $|i\rangle$ is the electronic wave function of the unexcited molecule. After the resonance interaction with the field (1) the wave function of the molecule will have the form

$$|\Psi, t\rangle = |i, t\rangle|\Phi_i, t\rangle - i \frac{(\mathbf{E}_0 \cdot \mathbf{d}_{i,e})}{2\hbar u} |e, t\rangle|V, t\rangle, \quad \frac{|\mathbf{E}_0 \cdot \mathbf{d}_{i,e}|}{2\hbar u} \ll 1. \quad (10)$$

Eq. (10) is derived in the first-order perturbation theory. Here $\mathbf{d}_{i,e}$, is the dipole matrix element of the electronic transition, which is assumed to be independent of the vibrational coordinate in accordance with the Condon approximation, and $|V, t\rangle$, the unnormalized vibrational wave function of the excited molecule. Due to (1) this wave function consists of two terms

$$|V, t\rangle = |V_+, t\rangle + \alpha |V_-, t\rangle, \quad (11)$$

The functions $|V_{\pm}, t\rangle$ correspond to the vibrational wave packets being excited respectively by each of the subpulses in (1). Generally, the wave functions $|V_+, t\rangle$ and $|V_-, t\rangle$ are not orthogonal and their overlap depends on the delay time T between the subpulses. This opens a possibility to observe the delay time dependent interference effects in the physical processes which are determined by the population of the excited electronic state of the molecule. For the sake of explicitness we shall assume that the measuring quantity is the quantum fluorescence yield of the excited molecule. In the case of the transition in the pure electronic system (without a vibrational degree of freedom) functions $|V_{\pm}, t\rangle$ are reduced to numerical amplitudes

$$V_{\pm} = 2^{-1/2} \pi^{-1/4} \exp\left[-\frac{\delta^2}{2u^2} \mp \frac{i\delta T}{2}\right], \quad (12)$$

where $\delta = \varepsilon_e/\hbar - \varepsilon_i/\hbar - \Omega$. The dependence of the excited state population on the delay time between the subpulses in this case is of a trigonometric character:

$$|V|^2 = 2\pi^{-1/2} \exp\left(-\frac{\delta^2}{u^2}\right) [1 + |\alpha|^2 + 2|\alpha|\cos(\varphi + \delta T)]. \quad (13)$$

In the remainder part of the paper we shall consider how the accounting of the molecular vibrations changes Eq. (12). The general expressions for functions $|V_{\pm}, t\rangle$ are given by

$$|V_{\pm}, t\rangle = \frac{u}{\pi^{1/4}} \int_{-\infty}^{\infty} dt_1 \exp\left[-\frac{u^2}{2}(t_1 \pm T/2) - i\Omega t_1 + \frac{i}{\hbar} H_e(t_1 - t) + \frac{i}{\hbar} \varepsilon_e t\right] \exp\left[\frac{i}{\hbar} H_i t_1\right] |\Phi_i\rangle. \quad (14)$$

Using (7) and (9) it is convenient to transform the wave functions (14) into the form of a distributed coherent state [13]

$$|V_{\pm}, t\rangle = \frac{u}{\pi^{1/4}} \exp\left(\mp i\delta \frac{T}{2}\right) \int_{-\infty}^{\infty} dt_1 \exp\left[-\frac{u^2}{2} t_1^2 + i\delta t_1\right] D\left(t_1 \mp \frac{T}{2} - t\right) |0\rangle. \quad (15)$$

Time dependence of the Heisenberg displacement operator $D(t)$ in (15) is determined by the Hamiltonian (6). To evaluate the time integral in (15) now it is possible to use the well known Fock-space expansion of the coherent state $D|0\rangle$ [23]. As a result we have:

$$|V_{\pm}, t\rangle = 2^{1/2} \pi^{-1/4} \sum_n \frac{(-g)^n}{\sqrt{n!}} \exp\left[-\frac{g^2}{2} - \frac{(\delta + n\omega)^2}{2u^2}\right] \exp\left[\mp \frac{i(\delta + n\omega)T}{2}\right] |n, t\rangle, \quad (16)$$

$$|n, t\rangle = \exp(-in\omega t) |n\rangle,$$

where $|n\rangle$ are the Fock-space eigenfunctions of the harmonic oscillator. Comparing Eqs. (12) and (16) it is easy to see that the coefficients at $|0, t\rangle$ in (16) coincide with the amplitudes V_{\pm} . Exact Eq. (16) is convenient for the numerical calculations. Another way to evaluate the integral (15) is based on the replacement of the coherent state $D\left(t_1 \mp \frac{T}{2} - t\right) |0\rangle = \left|-g e^{i\omega(t_1 \mp \frac{T}{2} - t)}\right\rangle$ by

$\left| -ge^{-i\omega(t \pm T/2)}(1 - i\omega t) \right\rangle$, which is possible when $u \gg \omega$. In [13,15] it was shown that the integration in (15) in this case can be performed in the operator form, and leads to the result:

$$|V_{\pm}, t\rangle \approx W^{-1/2} \exp\left(\mp i \frac{\delta T}{2}\right) \tilde{D}\left(t \pm \frac{T}{2}\right) S\left(t \pm \frac{T}{2}\right) |0\rangle. \quad (17)$$

In (17) the normalizing factor W is the absorption spectrum of the electronic-vibrational transition:

$$W = \pi^{-1/2} \frac{u}{B_0} \exp\left(-\frac{\Delta^2}{B_0^2}\right), \quad \Delta = \delta + g^2\omega, \quad B_0^2 = u^2 + 2g^2\omega^2, \quad (18)$$

\tilde{D} and S are the unitary displacement and squeezing operators:

$$\begin{aligned} \tilde{D}(t) &= \exp\left\{-g\left(1 - \frac{\omega \Delta}{B_0^2}\right)[b^+(t) - b(t)]\right\}, \\ S(t) &= \exp\left\{\frac{1}{2} \ln\left(\frac{B_0}{u}\right) [b(t)^2 - b^-(t)^2]\right\}. \end{aligned} \quad b(t) = \exp(-i\omega t)b, \quad (19)$$

The action on the vacuum state of the operator $\tilde{D}S$ gives an ideal coherent squeezed state in which, generally, the mean position and momentum of the oscillator have nonzero value, the uncertainties of the position and the momentum are not equal to each other and their product has the minimal possible value. In the a-plane of the coherent states each of the vectors $|V_{\pm}, t\rangle$ can be considered as an ellipse uniformly moving along the circle with the radius $\tilde{g} = g\left(1 - \frac{\omega \Delta}{B_0^2}\right)$, and keeping the orientation of the axes in the rotating frame. At moments t , for which the arguments of the operators in (17) are equal to the integer number of the vibrational periods, the values of the ellipse main axes are equal to the non-dimensional uncertainties of the position and the momentum with the small axis corresponding to the uncertainty of the position:

$$\Delta q = \frac{u}{\sqrt{2}\sqrt{2g^2\omega^2 + u^2}}, \quad \Delta p = \frac{\sqrt{2g^2\omega^2 + u^2}}{\sqrt{2}u}. \quad (20)$$

If the coupling constant is large enough ($g \gg 1$), then the wave functions $|V_{\pm}, t\rangle$ are located at a sufficient distance from each other, and are approximately orthogonal except for the case when the time delay between the subpulses appears to be equal to an integer number of the periods. From geometrical considerations it is clear that when the delay time between the subpulses gains an integer number of the vibrational periods, the overlap of the vibrational wave functions $|V_{+}, t\rangle$ and $|V_{-}, t\rangle$ can be observed in the a-plane as the approaching of two ellipses along the direction of their big axes. So it is likely to expect the following qualitative effect of the molecular vibrations on the interference picture given by Eq. (13): (i) the dependence of the population of an excited state on the delay time between the subpulses should be more pointed in comparison to the trigonometric dependence in Eq. (13), (ii) while the delay time approaches the integer number of the periods the interference picture should be sensitive to the quantum state of the molecular vibrations, and the squeezing effect can be seen due to the dependence of the interference picture on the subpulse duration.

Let us put $T = \frac{2\pi m}{\omega} + \tau$ in Eqs. (15), (16), where τ , is a small deviation from the integer number of the vibrational periods, and evaluate the population of the excited state. In accordance with Eqs. (15) and (16) one finds

$$\langle V|V\rangle = u \int_{-\infty}^{\infty} d\eta \exp\left(i\delta\eta - \frac{u^2}{4}\eta^2\right) \left[\begin{array}{c} (1+|\alpha|^2)M(\eta) + \\ \alpha e^{i\left(\frac{2\pi m}{\omega} + \tau\right)\delta} M(\eta + \tau) + \alpha^* e^{-i\left(\frac{2\pi m}{\omega} + \tau\right)\delta} M(\eta - \tau) \end{array} \right], \quad (21)$$

where $M(\xi) = \langle 0|D^+(0)D(\xi)|0\rangle$, and

$$\langle V|V\rangle = 2\pi^{1/2} \sum_n \frac{g^{2n}}{n!} \exp\left[-g^2 - \frac{(\delta + n\omega)^2}{u^2}\right] \left\{ 1 + |\alpha|^2 + 2|\alpha| \cos\left[\varphi + 2\pi m \frac{\delta}{\omega} + (\delta + n\omega)\tau\right] \right\}. \quad (22)$$

While deriving Eq. (21) we have performed the integration over the summary time $\frac{1}{2}(t_1 + t_1')$. The remaining integration in (21) is carried out over the difference time $t_1 - t_1' = \eta$. The vacuum average of two displacement operators in (21) frequently arises in the theory of Franck-Condon transitions and can be evaluated, for example, using the coherent states method [23]:

$$M(\xi) = \exp[g^2(e^{i\omega\xi} - 1)]. \quad (23)$$

It is worth noting that Eq. (22) can be received from (21) by expanding in (23) the exponent in the powers of $g^2 \exp(i\omega\xi)$. Due to the condition $u \gg \omega$ integral in (21) can be evaluated asymptotically (by the steepest descent method [24]). To do that one can expand the index of the exponent in the powers of ξ up to the second order terms:

$$M(\xi) = \exp\left(ig^2\omega\xi - \frac{1}{2}g^2\omega^2\xi^2\right). \quad (24)$$

Substituting (24) into (21) and performing the integration, one obtains

$$\langle V|V\rangle = W \left\{ 1 + |\alpha|^2 + 2|\alpha| \exp\left[-\frac{1}{4}\left(\frac{\tau}{\gamma_0}\right)^2\right] \cos\left(\varphi + \frac{2\pi m\delta}{\omega} + \frac{\Delta u^2 \tau}{B_0^2}\right) \right\}, \quad (25)$$

$$\gamma_0 = \frac{B_0}{\sqrt{2}g\omega u} = \frac{\sqrt{u^2 + 2g^2\omega^2}}{\sqrt{2}ug\omega}.$$

Equation (25) describes the interference between the vibrational wave packets within a single molecule. Depending on the experimental conditions, i.e. the values of φ , δ , and m , this interference can lead to a sharp peak (constructive interference), or to a down-fall (destructive interference) in the fluorescence dependence on the delay time between the subpulses. The new and the most constitutive feature of Eq. (25) is the dependence of the interference peak width γ_0 on the reciprocal pulse duration u . Within the range of pulse duration $\omega^2 \ll u^2 \ll 2g^2\omega^2$, where, in accordance with Eq. (20), the squeezing effect is the most considerable, the width of the interference peak approximately equals to the subpulse duration $\gamma_0 \approx u^{-1}$. This gives a practical possibility to use this effect for the duration measurement of a femtosecond laser pulse. The fact that the dependence of the interference peak width on the pulse duration is indeed the consequence of the dynamical squeezing effect becomes clear if we note that the second order expansion in Eq. (24) is equivalent to the first order expansion over $t_1 \pm \tau/2$ in Eq. (15). In the limit of extremely short pulses $u^2 \gg 2g^2\omega^2$ Eq. (25) coincides with that previously received in [20]. In that limit the

interference peak width does not depend on the pulse duration: $\gamma_0 \approx (\sqrt{2}g\omega)^{-1}$. It should be noted, that the limiting interference peak width is achieved for those laser pulses, during the action of which the initial vibrational state of a molecule in the process of electronic excitation has no time to change. The effect of the dynamical squeezing in this limit is absent. The picture of the interference peak as the function of the variables τ and u , numerically evaluated with the help of Eq. (22), can be seen in Fig. 1.

The case of nonzero temperature. In the case of nonzero temperature we shall assume that the initial vibrational state of the molecule is described by the equilibrium density matrix. The population of the excited electronic state in this case can be evaluated in the second order perturbation theory for the density matrix. An expression for the nonequilibrium density matrix of the phonons, arising due to the excitation of a Franck-Condon transition by a single laser pulse was obtained in [11, 14]. In the present paper we need to calculate the trace of an analogous density matrix, appearing due to the action of the field (1). It seems to be evident, that the expression for this trace will have the form of Eq. (21) with the functions $M(\xi)$ replaced by

$$\langle M(\xi) \rangle = \text{Sp}[\rho_0 D^+(0) D(\xi)] = \exp\{g^2[(\bar{n}+1)e^{i\omega\xi} + \bar{n}e^{-i\omega\xi} - 2\bar{n} - 1]\}, \quad (26)$$

where ρ_0 is the equilibrium density matrix of phonons, and \bar{n} , the thermal equilibrium phonon number. The Fourier expansion of $\langle M(\xi) \rangle$ is given by

$$\langle M(\xi) \rangle = \sum_{n=-\infty}^{+\infty} \left(\frac{\bar{n}+1}{\bar{n}}\right)^{\frac{n}{2}} I_n(2g^2\sqrt{\bar{n}(\bar{n}+1)}) \exp[-g^2(2\bar{n}+1) + in\omega\xi], \quad (27)$$

where I_n is the modified Bessel function. A generalization of equation (22) for the case of a nonzero temperature has the form

$$\langle V|V \rangle = 2\pi^{1/2} \sum_{n=-\infty}^{+\infty} \left(\frac{\bar{n}+1}{\bar{n}}\right)^{\frac{n}{2}} I_n(2g^2\sqrt{\bar{n}(\bar{n}+1)}) \exp\left[-g^2(2\bar{n}+1) - \frac{(\delta + n\omega)^2}{u^2}\right] \cdot \left\{1 + |\alpha|^2 + 2|\alpha| \cos\left[\varphi + 2\pi \frac{\delta}{\omega} + (\delta + n\omega)\tau\right]\right\}. \quad (28)$$

To evaluate the integral in Eqs. (21), in analogue to the case of the zero temperature, we shall expand the index of the exponent in Eq.(26) in the powers of ξ up to the second order terms. In this way we find that the effect of finite temperature in Eqs. (18) and (25) is manifested in the replacing coefficients B_0 and γ_0 by

$$B = \sqrt{u^2 + 2g^2\omega^2(2\bar{n}+1)}, \quad \gamma = \frac{\sqrt{u^2 + 2g^2\omega^2(2\bar{n}+1)}}{ug\omega\sqrt{2(2\bar{n}+1)}}. \quad (29)$$

From (29) one can see that the temperature growth leads to the extension of the area of linear dependence of the interference peak width on the laser pulse duration. According to. Eq. (29), $u_0^{-1} = [2g^2\omega^2(2\bar{n}+1)]^{-1/2}$ can be considered as the limiting value of pulse duration for the range of this linear dependence. It is interesting to estimate the value of u_0^{-1} for a real physical system. To do this we shall use the data for I_2 molecule from [22]. Taking $g^2 = 6.4$, $2\pi/\omega = 300$ fs and $T = 300^0$ K , we obtain $u_0^{-1} \approx 7$ fs. The natural upper limit for u_0^{-1} is the inverse vibrational frequency ω^{-1} . In the experiment [22] the laser pulses of about 50 fs duration have been used. In accordance with the given estimate the pulses of such duration belong to the area where $\gamma \approx u^{-1}$.

In conclusion note that the measurements of the interference peak width aimed at determining the duration of a femtosecond laser pulse represents only one of the possibilities to use the intramolecular interference phenomenon of the vibrational wave packets. Another interesting and important possibility is the observation of the wave packet distortions due to the intramolecular propagation. This problem we are planning to consider in our forthcoming paper.

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