Vibrational Schrödinger-cat states

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The optical Schrödinger-cat states are simple realizations of quantum states having nonclassical features. It is shown that vibrational analogs of such states can be realized in an experiment of doublepulse or chirped-pulse excitation of vibronic transitions. The possibility of conversion of vibrational Schrödinger-cat states to chemical-cat states is discussed.

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I. 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

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

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

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

Recently, great interest has been paid to such superposition states in quantum optics [1-8]. Nonclassical features of Schrödinger-cat states, i.e., squeezing [3], sub-Poissonian statistics, oscillation in photon statistics, etc., were discussed rather widely. It was shown [4,6] how the quantum interference between the coherent states involved in the superposition leads to the occurrence of nonclassical 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 Schrödinger-cat states [7,8].

A wide interest was addressed to wave-packet forma-

tion and motion during Franck-Condon transitions in both theoretical and experimental points of view [3,9,10]. In this paper, we shall discuss the possibilities of producing Schrödinger-cat-like superpositions of the vibrational states during Franck-Condon vibronic transitions in molecules or in crystals. As we shall see such states can be created by two short pulses separated in time or by appropriately chirped single pulses with comparatively long duration.

II. THE MODEL HAMILTONIAN

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

$$H_i = \epsilon_i + \frac{\hat{p}^2}{2M} + \frac{M\omega_i^2}{2}(\hat{q} + q_i)^2 , \qquad (2)$$

$$H_e = \epsilon_e + \frac{\hat{p}^2}{2M} + \frac{M\omega_e^2}{2}\hat{q}^2 , \qquad (3)$$

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,

$$\hat{q} = \left[\frac{\hbar}{2M\omega_e}\right]^{1/2} (b^{\dagger} + b), \quad \hat{p} = i \left[\frac{\hbar M\omega_e}{2}\right]^{1/2} (b^{\dagger} - b) , \qquad (4)$$

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

$$H_{i} = \epsilon_{i} + \frac{M\omega_{i}^{2}}{2}q_{i}^{2} + \hbar\omega_{i} \left\{ \frac{1}{4} \left[\frac{\omega_{i}}{\omega_{e}} + \frac{\omega_{e}}{\omega_{i}} \right] (b^{\dagger}b + bb^{\dagger}) + \frac{1}{4} \left[\frac{\omega_{i}}{\omega_{e}} - \frac{\omega_{e}}{\omega_{i}} \right] (b^{\dagger}b^{\dagger} + bb) \right\} + \frac{\hbar\omega_{i}^{2}q_{i}}{\omega_{e}} \left[\frac{M\omega_{e}}{2\hbar} \right]^{1/2} (b^{\dagger} + b) ,$$
(5)

$$H_e = \epsilon_e + \frac{1}{2} \hbar \omega_e (b^{\dagger} b + b b^{\dagger}) .$$
(6)

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

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

$$g = q_i \left(\frac{M\omega_e}{2\hbar}\right)^{1/2}, \quad r = \frac{1}{2} \ln \frac{\omega_i}{\omega_e}.$$

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

$$|0\rangle_{i} = \hat{\Sigma}|0\rangle_{e} , \qquad (8)$$

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

The Hamiltonian H'(t) describing the interaction with the external field has the form

$$H'(t) = \frac{1}{2} d_{ie} E(t) a_e^{\dagger} a_i + \frac{1}{2} d_{ie}^{*} E^{*}(t) a_i^{\dagger} a_e \quad , \tag{9}$$

where

$$E(t) = e(t)\exp(-i\Omega_0 t) , \qquad (10)$$

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

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

$$|\Psi,t\rangle = |i,t\rangle|0\rangle_{i} - i\frac{E_{0}d_{ie}}{2\hbar}|e,t\rangle|\{E(t)\}\rangle_{e} .$$
(11)

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

$$\left|\left\{E\left(t\right)\right\}\right\rangle_{e} = \int_{-\infty}^{\infty} d\tau E\left(\tau\right) \exp\left[\frac{i}{\hbar}H_{e}(\tau-t) + \frac{i}{\hbar}\epsilon_{e}t\right]$$
$$\times \exp\left[-\frac{i}{\hbar}H_{i}(\tau-t)\right]\left|0\right\rangle_{i}.$$
 (12)

Applying Eqs. (7), (8) the wave function of the excited state Eq. (12) can be transformed to the following form

$$|\{E(t)\}\rangle_{e} = \int_{-\infty}^{\infty} d\tau \exp(i\delta\tau) e(\tau) \widehat{\Sigma}(\tau-t) |0\rangle_{e} , \qquad (13)$$

where $\delta = \overline{\Omega} + (\omega_e - \omega_i)/2 - \Omega_0$, $\overline{\Omega} = (\epsilon_e - \epsilon_i)/\hbar$.

In the following sections we shall investigate the properties of the vibrational wave function of Eq. (13) considering different types of 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

$$\{E(t)\}\rangle_e = \int_{-\infty}^{\infty} d\tau \exp(i\delta\tau) e(\tau) |\alpha(\tau-t)\rangle_{\rm coh} , \qquad (14)$$

where $|\alpha(\tau-t)\rangle_{\rm coh}$ is a coherent state with respect to the phonon operator b and $\alpha(\tau-t)=ge^{-i\omega(\tau-t)}$.

III. DOUBLE-PULSE EXCITATION

In the most simple case the exciting pulse is transform limited with a Gaussian shape

$$E(t) = E_0 \pi^{-1/4} e^{-(u^2/2)t^2 - i\Omega_0 t}, \qquad (15)$$

where u is the reciprocal pulse duration. The properties of the emerging wave function were studied in Refs. [3], [11]. This wave function, depending on the pulse duration, corresponds to several of the most important states in quantum optics. In the case of extremely short pulses the wave function describes a usual coherent state [Fig. 1(a)], while in the opposite limit of long pulses it is the *n*phonon number state [Fig. 1(b)]. Between these limiting cases it is close to a squeezed minimal uncertainty state [Fig. 1(c)], or, for longer pulses it is the bananalike amplitude squeezed state [Fig. 1(d)], which also appears to be an approximate number-phase intelligent state [12] associated with Pegg-Barnett phase operator formalism [13]. The vibrational squeezing predicted theoretically in Refs. [3], [10] has been experimentally found recently [14].

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

$$E(t) = \frac{E_0}{2\pi^{1/4}} e^{-(u^2/2)[t + (T_1/2)]^2 - i\Omega_0[t + (T_1/2)]} + \frac{E_0}{2\pi^{1/4}} e^{-(u^2/2)[t - (T_1/2)]^2 - i\Omega_0[t - (T_1/2)] - i\phi},$$
(16)

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

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

$$\{u, T_1, \delta, \phi\} \rangle_e = e^{-i\overline{\Omega}(T_1/2)} \left| u, \left| t + \frac{T_1}{2} \right| \right) + e^{i\overline{\Omega}(T_1/2) - i\phi} \left| u, \left| t - \frac{T_1}{2} \right| \right), \quad (17)$$

$$|u,t\rangle = \frac{u}{\pi^{1/4}} \int_{-\infty}^{\infty} d\tau \, e^{-(u^2/2)\tau^2 + i\delta\tau} |\alpha(\tau - t)\rangle_{\rm coh} \,.$$
(18)

To investigate the quantum properties of the superposition state of Eq. (17) it is convenient to consider its Wigner function

$$W(\alpha) = \frac{1}{\pi^2} \int e^{\eta^* \alpha - \eta \alpha^* - (1/2)|\eta|_e^2} \langle \{u, T, \phi\} | e^{\eta b^\dagger} e^{-\eta^* b} | \{u, T, \phi\} \rangle_e d^2 \eta .$$
⁽¹⁹⁾



FIG. 1. The time dependence of the absolute square of the wave function $|\psi(q)|^2$ of the excited vibrational state, produced by a single Gaussian-shaped laser pulse, during half vibrational period. Here x and t represent the position and the time, respectively. In (a), in the case of extremely short pulses $u \gg \omega$, one can see how a vibrational coherent state oscillates along a classical trajectory. For a slightly longer pulse the emerging vibrational state is practically a squeezed state as seen in (b). In the case of even longer pulses the vibrational state turns into a banana state (c) that evolves into the time independent *n*-phonon state for long pulses (d).

For extremely short pulses we have coherent superposition states which are the vibrational analog of the socalled optical Schrödinger-cat states. The Wigner function and the time dependence of the absolute square of the wave function are shown in Fig. 2(a) and 2(b) 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.

For long, strongly overlapping pulses Eq. (17) leads to the phonon-number state shown in Fig. 1(d).

In the intermediate case of short (typically femtosecond) pulses the Wigner function of this state consists of symmetrically situated bananalike shapes revolving clockwise with the vibrational frequency along a circle with a radius of the shift parameter g. A wavelike fringe pattern, according to the interference, spins between them with the same frequency ω [Fig. 3(a)]. The graph of the absolute square of the wave function in Fig. 3 shows that if the bananalike shapes of the Wigner function have different projections on the real axis the fringes practically disappear after the integration over the imaginary axis. For some time intervals $\Delta t(u)$ around $t = (n + \frac{1}{2})T/2$ (n is 0 or positive integer), the fringes survive the integration. As the duration of the subpulses increases, this interval becomes longer [Fig. 3(b)], resulting in the number state [Fig. 1(d)].

For small coupling constants $(g < \omega)$ the bell-like shapes are not resolved even for ultrashort pulses. In this case, the quantum interference results not in oscillations but in narrowing in the spatial distribution in some intervals of time in every half a period of the vibration.

IV. CHIRPED-PULSE EXCITATION

Let us consider a single exciting pulse with linear chirp and Gaussian envelope function,

$$E(t) = \frac{E_0}{2\pi^{1/4}} e^{-(u^2/2)t^2 + i(w^2/2)t^2 - i\Omega_0 t} .$$
 (20)

Depending on the actual values of the pulse duration (u^{-1}) and chirp (w) the effective pulse can correspond to different type of usual pulses from single ultrashort pulses to the double pulses of the previous section. A convenient method to see what kind of effective pulses we have is the investigation of the spatial uncertainty ΔX_{+}^{2} of the emerging excited vibrational state. In Fig. 4, the dependence of the spatial uncertainty arising from the chirp is shown for a relatively long exciting pulse with reciprocal pulse duration $u = 0.1\omega$. Typical value of the uncertainty ΔX_{+}^{2} in Fig. 4, is about 19 corresponding to the uncertainty of the number state $\Delta X_{+}^{2} = 2n+1$, $n = (g/\omega)^2 = 9$. This indicates that the resulting vibrational state is similar to a Fock state and more or less equally distributed on the circle of radius g on the α plane. The maxima and minima of the curve have a more



FIG. 2. The Wigner function (a) and the time dependence of the absolute square of the wave function $|\psi(q)|^2$ (b) of the Schrödingercat 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 $t = \pm T/4$, otherwise in the bigger part of the period $|\psi(q)|^2$ consists of two Gaussians representing the two superposed coherent states.

interesting origin. A minimal value of the uncertainty ΔX_{+}^2 indicates that the state has a narrow spatial distribution, if it is less than unity the state is squeezed [Fig. 5(a)]. On the other hand, a state with maximal value of ΔX_{+}^2 , i.e., with a large second momentum, is symmetrically distributed on the circle as faraway as possible from the imaginary axis [Fig. 5(b)]. The state shown in Fig. 5(b) is in fact a Schrödinger-cat state produced by chirped single-pulse excitation.

The emerging of such states during comparatively long pulse excitation can be explained as follows. Suppose that the duration of the exciting pulse covers several vibrational periods. Due to the periodicity of the vibration the effect from the electric field $E_1 = E(t_1)$ at time t_1 will be the same as if the same electric field E_1 was applied at $t_1 \pm T$. Therefore, the laser pulse E(t) can be substituted by an effective "reduced pulse" $\tilde{E}(t)$ which is confined within one vibrational period



FIG. 3. The time dependence of the absolute square of the wave function $|\psi(q)|^2$ of the vibrational superposition state formed by double-pulse electronic-vibrational excitation. Ultrashort pulses form a vibrational state shown in (b), while a little longer pulses form a squeezed state superposition (a) or banana state superposition (b) states. The oscillation area around $t = \pm T/4$ becomes longer as the exciting pulse duration increases.

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$$\widetilde{E}(t) = \frac{E_0}{2\pi^{1/4}} \sum_{j=-\infty}^{\infty} e^{-(u^2/2)(t+jT)^2 + i(w^2/2)(t+jT)^2 - i\Omega_0(t+jT)}, \quad -\frac{T}{2} < t < \frac{T}{2}$$
(21)

Such an effective pulse $\tilde{E}(t)$ is shown in Fig. 6, as a function of chirp of the original pulse for reciprocal pulse duration $u = 0.1\omega$. As it can be seen, there are values of the chirp $w \approx 0.399\omega$ and $w \approx 0.564\omega$ for which the effective pulse is single peaked marked by 1 [leading to squeezed or banana-shaped states shown in Fig. 5(a)] or double peaked marked by 2 [resulting in Schrödinger-cat-like states shown in Fig. 5(b)].

V. POPULATION ANALYSIS

Let us consider the excitation spectrum of the transition from the initial to the excited electronic level introduced by double-pulse excitation. The population of the excited level as a function of the excitation central frequency is proportional to the norm of the unnormalized vibrational wave function of Eqs. (17), (18)

$$= \frac{u^{2}}{\pi^{1/2}} \int_{-\infty}^{\infty} dt_{1} \int_{-\infty}^{\infty} dt_{2} e^{i\delta(t_{1}-t_{2})+g^{2}[e^{i\omega(t_{1}-t_{2})}-1]}} \times \{e^{-(u^{2}/2)[t_{1}+(T_{1}/2)]^{2}-(u^{2}/2)[t_{1}+(T_{1}/2)]^{2}} + e^{-(u^{2}/2)[t_{2}+(T_{1}/2)]^{2}-(u^{2}/2)[t_{1}-(T_{1}/2)]^{2}-i\phi} + e^{-(u^{2}/2)[t_{2}+(T_{1}/2)]^{2}-(u^{2}/2)[t_{1}-(T_{1}/2)]^{2}-(u^{2}/2)[t_{2}-(T_{1}/2)]^{2}}\}.$$
 (22)

Substituting new integral variables $\xi = t_1 - t_2$ and $2\tau = t_1 + t_2$ into Eq. (22) and evaluating the integral by τ we have

$$e^{\langle \{u, T_{1}, \delta, \phi\} | \{u, T_{1}, \delta, \phi\} \rangle_{e}}$$

= $u \int_{-\infty}^{\infty} d\xi e^{i\delta\xi + g^{2}[e^{i\omega\xi} - 1]}$
 $\times \{2e^{-(u^{2}/4)\xi^{2}} - e^{-(u^{2}/4)(\xi - T_{1})^{2} - i\phi}$
 $+ e^{-(u^{2}/4)(\xi + T_{1})^{2} - i\phi} \}.$ (23)



FIG. 4. The variance of the quadrature operator ΔX_{+}^2 at different values of the exciting pulse chirp for a pulse with reciprocal duration $u = 0.1\omega$ (the coupling constant of the electronic-vibrational transition $g = 3\omega$). The minimal (1) and maximal (2) values of ΔX_{+}^2 are related to states that, instead of longer chirped pulses could have been formed by short transform limited single (1) or double (2) pulses correspondingly.

Finally, substituting the expression $\exp(g^2 e^{i\omega\xi})$ by its series

$$\exp(g^2 e^{i\omega\xi}) = \sum_{n=1}^{\infty} \frac{g^{2n}}{n!} e^{i\omega n\xi} , \qquad (24)$$

and evaluating the second integral we find

$$e^{\langle \{u, T_1, \delta, \phi\} | \{u, T_1, \delta, \phi\} \rangle_e}$$

= $4\sqrt{\pi}e^{-g^2} \sum_n \frac{g^{2n}}{n!} e^{-(\delta + n\omega)^2/u^2}$
 $\times \{1 + \cos[\phi + (\delta + n\omega)T_1]\}.$ (25)

For the limit of vanishing g=0, Eq. (25) simplifies to

$$\langle \{u, T_1, \delta, \phi\} | \{u, T_1, \delta, \phi\} \rangle_e$$

=4 $\sqrt{\pi}e^{-\delta^2/u^2} \{1 + \cos[\phi + \delta T_1]\}$, (26)

while the opposite limit of large g and very short pulses leads to a periodic solution with a period of the molecular vibrations. The excited-state population has a maximum (for $\phi = 0$) or a minimum (for $\phi = \pi$) if the delay between

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FIG. 5. Wigner functions of vibrational states produced by comparatively long (reciprocal pulse duration $u = 0.1\omega$) pulses with linear chirps $w \approx 0.399\omega$ (a) and $w \sim 0.564\omega$ (b). (a) and (b) correspond to the minimal and maximal values of the quadrature operator uncertainty ΔX_{+}^2 marked as (1) and (2), respectively.



the pulses is a multiple of the vibrational period (i.e., $T_1 = nT$, *n* being a positive integer). The explanation is similar to the considerations in Sec. IV resulting in introducing the effective pulse $\tilde{E}(t)$ for chirped pulses: due to periodicity an electric field at some moment or a period later has the same impact on the molecule's vibration. This effect for ultrashort pulses was used to conceive a wave-packet interferometry [16,15].

Equation (26) for the case of g=0 reflects the fact that in the Fourier spectrum of a double pulse there are periodical downfalls.

VI. DISCUSSION

In Sec. III it is shown that temporary superposition of the exciting pulse can be converted into a quantummechanical superposition state of the vibrational mode of the laser excited molecule. We have analyzed how chirped pulse excitation can lead to creation of well separated quantum superposition due to constructive and destructive interferences of the molecular vibration.

By a secondary excitation using probe pulse(s) one may



FIG. 7. 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 "Schrödinger-cat" state.

transfer the vibrational cat state into chemical cat state creating quantum superposition of different molecules [17]. Baumert *et al.* excited the Na₂ molecule by a short laser pulse. 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

- [1] B. Yurke and D. Stoler, Phys. Rev. Lett. 57, 13 (1986).
- M. Hillery, Phys. Rev. A 36, 3796 (1987); Y. Xia and G. Guo, Phys. Lett. A 136, 281 (1989); G. C. Gerry, Opt. Commun. 91, 47 (1992).
- [3] J. Janszky and An. V. Vinogradov, Phys. Rev. Lett. 64, 2771 (1990).
- [4] W. Schleich, M. Pernigo, and Fam Le Kien, Phys. Rev. A 44, 2172 (1991); V. Buzek and P. L. Knight, Opt. Commun. 81, 331 (1991); Jinzuo Sun, Jisuo Wang, and Chuankui Wang, Phys. Rev. A 46, 1700 (1992).
- [5] M. S. Kim and V. Buzek, Phys. Rev. A 46, 4239 (1992), 47, 610 (1993); I. Jex and V. Buzek, J. Mod. Opt. 40, 771 (1993).
- [6] J. Janszky, P. Domokos, and P. Adam, Phys. Rev. A 48, 2213 (1993); P. Adam, I. Foldesi, and J. Janszky, *ibid.* 49, 1281 (1994).
- M. Brune, S. Haroche, V. Lefevre, J. M. Raimond, and N. Zagury, Phys. Rev. Lett. 65, 976 (1990); M. Brune, S. Haroche, J. M. Raimond, L. Davidovich, and N. Zagury, Phys. Rev. A 45, 5193 (1992).
- [8] T. Ogawa, M. Ueda, and N. Imoto, Phys. Rev. Lett. 66, 1046 (1991); Phys. Rev. A 43, 6458 (1991); B. Sherman and G. Kurizki, *ibid.* 45, 7674 (1992).
- [9] An. V. Vinogradov and J. Janszky, Fiz. Tverd. Tela (Leningrad) 27, 892 (1985) [Sov. Phys. Solid State 27, 598 (1985)]; D. J. Tannor and S. A. Rice, J. Chem. Phys. 83, 5013 (1985); M. J. Rosker, F. W. Wise, and C. L. Tang,

another excited level or dissociated fragments. We suggest a similar experiment with a double-pulse primary excitation leading to a Schrödinger vibrational state on the level e (Fig. 7). Applying a third pulse when the two parts of the Schrödinger-cat state are furthest from each other one obtains a superposition of a molecule with its fragment. If we demand that the molecular and electronic state be separable, i.e., the state of the molecule be pure, it is necessary that either the Fourier spectra of the transform limited third pulse covers the energies of the two Franck-Condon transitions ($e \rightarrow A$, $e \rightarrow B$ in Fig. 7) or the third exciting pulse has two central frequencies phase locked to each other.

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

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Phys. Rev. Lett. 57, 321 (1986); P. Kowalczyk, C. Radzewicz, J. Mostowski, and I. A. Walmsley, Phys. Rev. A 42, 5622 (1990); M. Gruebele and A. Zewail, Phys. Today 43 (5), 24 (1990); W. T. Pollard, S.-Y. Lee, and R. A. Mathies, J. Chem. Phys. 92, 4012 (1990); E. D. Potter, J. L. Herek, S. Pedersen, Q. Liu, and A. H. Zewail, Nature 355, 66 (1992); G. M. Garraway and S. Stenholm, Phys. Rev. A 46, 1413 (1992); T. Szakacs, J. Somloi, and A. Lorincz, Chem. Phys. 172, 1 (1993); I. Averbukh and M. Saphiro, Phys. Rev. A 47, 5086 (1993).

- [10] J. Janszky, T. Kobayashi, and An. V. Vinogradov, Opt. Commun. 76, 30 (1990).
- [11] An. V. Vinogradov and J. Janszky, Zh. Eksp. Teor. Fiz.
 99, 386 (1991) [Sov. Phys. JETP 73, 211 (1991)]; J. Janszky, P. Adam, An. V. Vinogradov, and T. Kobayashi, Spectrochim. Acta, Part A 48, 31 (1992).
- [12] P. Adam, J. Janszky, and An. V. Vinogradov, Phys. Lett. A 160, 506 (1991).
- [13] D. T. Pegg and S. M. Barnett, Phys. Rev. A 39, 1665 (1989).
- [14] T. J. Dunn, J. N. Sweetser, J. A. Walmsley, and C. Radewicz, Phys. Rev. Lett. 70, 3388 (1993).
- [15] N. F. Scherer, R. J. Carlson, A. Matro, Mei Du, A. J. Ruggiero, V. Romero-Rochin, J. A. Cina, G. R. Fleming, and S. A. Rice, J. Chem. Phys. 95, 1487 (1991).
- [16] E. Tokunaga, A. Terasaki, and T. Kobayashi, Opt. Lett.
 17, 1133 (1992); 18, 370 (1993); Phys. Rev. A 47, R4581

(1993).

 T. Baumert, M. Grosser, R. Thalweiser, and G. Gerber, Phys. Rev. Lett. 67, 3753 (1991); T. Baumert, R. Thalweiser, V. Weiss, and G. Gerber, in Ultrafast Phenomena VIII, edited by J.-L. Martin, A. Migus, G. A. Mourou, and A. H. Zewail, Springer Series in Chemical Physics Vol. 55 (Springer-Verlag, Berlin, 1993); M. S. Kurdoglyan (private communication).



FIG. 2. The Wigner function (a) and the time dependence of the absolute square of the wave function $|\psi(q)|^2$ (b) of the Schrödingercat 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 $t = \pm T/4$, otherwise in the bigger part of the period $|\psi(q)|^2$ consists of two Gaussians representing the two superposed coherent states.



FIG. 5. Wigner functions of vibrational states produced by comparatively long (reciprocal pulse duration $u = 0.1\omega$) pulses with linear chirps $w \approx 0.399\omega$ (a) and $w \sim 0.564\omega$ (b). (a) and (b) correspond to the minimal and maximal values of the quadrature operator uncertainty ΔX_{+}^2 marked as (1) and (2), respectively.



FIG. 6. The reduced effective electric field $\tilde{E}(t)$ as a function of the chirp parameter w. Here, as an example, the reciprocal pulse duration $u = 0.1\omega$ and the central frequency of the pulse is supposed to be $\Omega_0 = (n + \frac{1}{3})\omega$, where ω is the vibrational frequency and n a positive integer. One can see that at some value of the chirp parameter marked by 1 the reduced electric field has a well pronounced peak, leading to a squeezed vibrational state shown in (a), while at another value of the chirp marked by 2 a double peaked $\tilde{E}(t)$ produces Schrödinger-cats-like vibrational states [see Fig. 5(b)].