Squeezing via One-Dimensional Distribution of Coherent States

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It is found that strong squeezing can be obtained by special superposition of coherent states along a straight line in the α plane. This mechanism opens new possibilities for squeezing, e.g., of the molecular vibrations during a Franck-Condon transition induced by a short coherent light pulse.

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Recently, a lot of interest was paid to the idea of squeezing[1,2] from both fundamental and practical points of view. The majority of the physical proposals for obtaining squeezed states is based on the employment of some two-photon process. In Ref. 3 an interesting new possibility for squeezing was suggested, namely, the superposition of the vacuum with the one- and two-photon state. In this paper we will show that an effective squeezing can be achieved by superposition of coherent states along a straight line on the α plane. (For this aspect of the graphic representation of squeezed states, see Ref. 4). We shall also give an example of a physical process leading to such a superposition.

First, let us consider superposition states defined by

$$|\alpha,\pm\rangle \equiv c_\pm(|\alpha\rangle \pm |-\alpha\rangle), \quad c_\pm = \{2[1\pm\exp(-2|\alpha|^2)]\}^{-1/2}. \tag{1}$$

$$b|\alpha,\pm\rangle = \alpha c_\mp^{-1} c_\pm |\alpha,\pm\rangle, \quad b^2|\alpha,\pm\rangle = \alpha^2|\alpha,\pm\rangle, \quad \langle\alpha,+|\alpha',-\rangle = 0, \tag{2}$$

where $|\alpha\rangle$ is the usual coherent state $b|\alpha\rangle = \alpha|\alpha\rangle$ and b is the annihilation operator. Similar states have been discussed recently in Refs. 5–9. For the sake of simplicity let us suppose that $\alpha = x$ is real. We can see that $|x,+\rangle$ is squeezed,

$$\Delta b_2^2 = \tfrac{1}{4} - x^2/[1+\exp(2x^2)], \tag{3}$$

where b_1 and b_2 are the Hermitian quadratures of the annihilation operator $b = b_1 + ib_2$. The maximum squeezing is at $x = 0.80$ where for the variance Δb_2^2 we find 0.111 instead of the corresponding vacuum value 0.25. The other state $|x,-\rangle$ does not show any squeezing though it is antibunched. The squeezing can be further enhanced if $|x,+\rangle$ is superposed with the vacuum state

$$|x,p\rangle \equiv [2+2\exp(-2x^2)+4p\exp(-x^2/2)+p^2]^{-1/2}(|x\rangle + p|0\rangle + |-x\rangle), \tag{4}$$

with a minimum value of $\Delta b_2^2 = 0.0651$ at $x = 1.57$ and $p = 1.35$.

An even higher level of squeezing can be obtained by the generalization of (1) and (4),

$$|\{F(x)\}\rangle \equiv c_F \int_{-\infty}^{\infty} F(x)|x\rangle dx, \tag{5}$$

$$c_F^{-2} = \int\int_{-\infty}^{\infty} F(x)F(x')\exp[-(x-x')^2/2]\,dx\,dx', \tag{6}$$

where, based on the analogy with (1) and (4), we supposed that $F(x)$ is a positive and even function. It is also assumed that the integrals

$$\langle (b^\dagger)^m b^n\rangle = c_F^2 \int\int_{-\infty}^{\infty} F(x)F(x')\exp[-(x-x')^2/2]x^m x'^n\,dx\,dx' \tag{7}$$

exist. Comparing Eq. (5) with Glauber's well-known expansion[10]

$$|f\rangle = \frac{1}{\pi}\int |\alpha\rangle f(\alpha^*)e^{-(1/2)|\alpha|^2}d^2\alpha, \tag{8}$$

where $f(\alpha^*)$ is an analytical function of α^*, we note that (5) is not a particular case of (8) because there is no analytical function $f(\alpha^*)$ leading from (8) to (5). On the other hand, Eq. (5) can be considered as a special case of the complex P representation.[11] We shall see that in expanding Eq. (5) such important nonclassical states as the squeezed and the amplitude squeezed states have an especially simple form.

For the variances of the quadrature b_2 from (5) after symmetrization over the integral variables we obtain

$$\Delta b_2^2 = 0.25 - 0.25 c_F^2 \int\int_{-\infty}^{\infty} F(x)F(x')\exp[-(x-x')^2/2](x-x')^2 dx\,dx' . \tag{9}$$

We can see that for any positive even function $F(x)$, integrable in the sense of (7), the state defined by (5) is squeezed except for $F(x)=\delta(x)$ describing the vacuum state.

Special choices of the distribution function $F(x)$ give us the states of expressions (1) and (4). Another special choice is the Gaussian distribution

$$F(x) = \pi^{-1/2}\gamma^{-1}(1+\gamma^2)^{1/4}\exp(-x^2\gamma^2), \quad c_F = 1 . \tag{10}$$

For the investigation of the statistical properties of our distributed coherent state it is convenient to find its characteristic function

$$\chi(\eta) = \mathrm{Tr}\{\rho\exp(\eta b^\dagger - \eta^* b)\} = c_F^2 \int\int_{-\infty}^{\infty} dx\,dx' F(x)F(x')\exp[-\tfrac{1}{2}|\eta|^2 + \eta x' - \eta^* x - (x-x')^2/2] , \tag{11}$$

$\eta = \mu + i\nu$. For a rather general class of states with coherent, chaotic, and squeezed features this characteristic function has a Gaussian form[12]

$$\chi(\eta) = \exp(-M\mu^2 - K\nu^2 + G\mu\nu - 2iP\mu + 2iQ\nu) ,$$
$$\Delta b_1^2 = K/2, \quad \Delta b_2^2 = M/2 , \tag{12}$$

where $P+iQ=W$ is the coherent signal. The squeezing is absent when $M=K$ and $G=0$. For pure squeezed states we have

$$4MK - G = 1 . \tag{13}$$

In the case of the state defined by (5) and (10) the characteristic function has a simple form

$$\chi(\eta) = \exp\left[-\frac{1}{1(1+\gamma^2)}\mu^2 - \frac{1+\gamma^2}{2}\nu^2\right] . \tag{14}$$

This characteristic function is a special case of Eq. (12). Satisfying condition (13) it describes a pure squeezed vacuum state with variances

$$\Delta b_1^2 = \frac{1+\gamma^2}{4}, \quad \Delta b_2^2 = \frac{1}{4(1+\gamma^2)} . \tag{15}$$

Thus we showed that a Gaussian distribution of coherent states along a straight line is a minimum uncertainty state, and for $\gamma^2 \gg 1$ manifests strong squeezing.

Equation (5) can be generalized in an obvious way by taking the distribution not along the real axis but along any straight line. Furthermore, one can construct a one-dimensional distribution along any contour in the α plane. For example, using a distribution along an arc

$$|\{f(\psi)\}\rangle \equiv c_f \int_{-\pi}^{\pi} d\psi f(\psi)\,|\alpha_0 e^{i\psi}\rangle , \tag{16}$$

one can model the amplitude squeezed state.[13]

The significance of the integral representation proposed in this paper is connected with the possibility of finding new ways of squeezed-state generation and also with a new aspect of understanding the nature of squeezing. It simplifies some calculations with squeezed states and allows one to treat non-Gaussian squeezed states as well.

Let us now turn to a physical example of the distributed coherent states. The problem we will consider is a Franck-Condon transition in a molecule induced by a short coherent light pulse. For the sake of simplicity we shall give the solution of this problem for a two-level one-mode electron-vibrational system. Let $|i\rangle$ be the ground state and $|j\rangle$ be the excited electronic state of the molecule. Suppose that due to the electronic transition $i \rightarrow j$ there is only a shift in the harmonic vibrational potential and no frequency change. Our aim is to study the vibrational state properties of the electronically excited state. For that reason it is convenient to consider the molecular vibrations in the variables connected with the excited state. Using these variables we can write the adiabatic Hamiltonians for each electronic state as

$$H_i = \tilde{\epsilon}_i + \hbar\omega b^\dagger b + \hbar\omega g(b^\dagger + b) , \tag{17}$$

$$H_j = \epsilon_j + \hbar\omega b^\dagger b . \tag{18}$$

Here $\tilde{\epsilon}_i$ and ϵ_j are the electronic energy levels, ω is the frequency of the vibration, $b^\dagger$ (b) is the creation (annihilation) phonon operator, and the interaction constant g is the ratio of the above-mentioned potential shift to the amplitude of the zero vibrations.

The Hamiltonian (17) can be diagonalized by the well-known displacement operator $D = \exp[-g(b^\dagger - b)]$,

$$D^\dagger H_i D = \epsilon_i + \hbar\omega b^\dagger b, \quad \epsilon_i = \tilde{\epsilon}_i - \tfrac{1}{2}g^2\hbar\omega . \tag{19}$$

From (19) it can be seen that the ground-state vibrational wave function of (17) is a coherent state $|-g\rangle = D|0\rangle$, where $|0\rangle$ is the phonon vacuum state. This simply reflects the fact that the ground-state wave function of a harmonic oscillator in respect to a shifted potential looks like a coherent state.

The whole Hamiltonian, which also includes the resonant interaction with the external classical electromagnetic pulse of field amplitude E_0, center frequency Ω, and reciprocal duration u, has the form

$$H=H_i a_i^\dagger a_i+H_j a_j^\dagger a_j+\mu_{ji}(t)a_j^\dagger a_i+\mu_{ij}(t)a_i^\dagger a_j\,,$$
$$\mu_{ji}(t)=-\frac{1}{2}d_{ji}\frac{E_0}{\pi^{1/4}}\exp\left[-i\Omega t-\frac{\mu^2}{2}t^2\right], \tag{20}$$
$$\mu_{ij}(t)=\mu_{ji}^*(t)\,,$$

where $a^\dagger$ (a) is the creation (annihilation) electron operator and d_{ji} is the dipole matrix element for the electronic transition $i\rightarrow j$.

Let us suppose that initially at $t=-\infty$ the system was in the ground state $|i\rangle|-g\rangle$. Then for times $t\gg u^{-1}$, using the first-order perturbation theory we obtain the Schrödinger wave function of the whole system

$$|\Psi,t\rangle=|i,t\rangle|-g\rangle+i\frac{d_{ij}E_0}{2\hbar\pi^{1/4}}|j,t\rangle|u,t\rangle\,,$$
$$\frac{|d_{ij}E_0|}{\hbar u}\ll 1\,, \tag{21}$$

where $|i,t\rangle=|i\rangle\exp(-i\epsilon_i t/\hbar)$ and the vibrational wave function of the electronically excited molecule has the form of the unnormalized distributed coherent state

$$|u,t\rangle=\int_{-\infty}^{\infty}d\tau\exp[-i\delta\tau-(u^2/2)\tau^2]\,|\alpha(t-\tau)\rangle\,,$$
$$\alpha(t-\tau)=-g\exp[-i\omega(t-\tau)],\quad \delta=\Omega-(\epsilon_j-\epsilon_i)/\hbar\,. \tag{22}$$

One can see that in (22) the coherent state is distributed along an arc with the spread depending on the duration of the exciting pulse. For extremely short pulses ($u\rightarrow\infty$) this distribution contracts into the usual coherent state. In this case $\Delta b_1^2=\Delta b_2^2=\frac{1}{4}$. For long pulses ($u\ll\omega$), having an equal distribution along the circle, (22) turns into the n-photon number state ($n\omega=\delta$) with $\Delta b_1^2=\Delta b_2^2=\frac{1}{4}+n/2$. Between these two limits the uncertainties do not necessarily change monotonously from $\frac{1}{4}$ to $\frac{1}{4}+n/2$ while the excitation pulse duration becomes longer and longer. Graphically it can be understood if one visualizes how the muffinlike coherent state going through a squeezed crescentlike shape deforms along a circle into the donutlike number state.[13]

Under condition $u\gg\omega$ only a small part of the arc contributes to the integral (22) and can be replaced by a straight segment

$$|\alpha(t-\tau)\rangle\approx|1-g\exp(-i\omega t)(1+i\omega\tau)\rangle\,. \tag{23}$$

Substituting (23) into (22) we have a state of the type (5). To find its squeezing properties let us calculate the characteristic function (11) for

$$\rho_u=|u,t\rangle\langle u,t|/\langle u,t|u,t\rangle\,.$$

After straightforward calculations we find the expression of form (12) with time-dependent coefficients

$$M(t)=\frac{1}{2}-\frac{g^2\omega^2}{2g^2\omega^2+u^2}\sin^2(\omega t)+\frac{g^2\omega^2}{u^2}\cos^2(\omega t)\,,$$
$$K(t)=\frac{1}{2}-\frac{g^2\omega^2}{2g^2\omega^2+u^2}\cos^2(\omega,t)+\frac{g^2\omega^2}{u^2}\sin^2(\omega t)\,,$$
$$G(t)=[M(0)-K(0)]\sin(2\omega t)\,, \tag{24}$$
$$Q(t)=-g\left[1-\frac{g^2\omega^2-\omega\delta}{2g^2\omega^2+u^2}\right]\cos(\omega t)\,,$$
$$P(t)=-Q(0)\sin(\omega t)\,,$$

which obey condition (13) for all t.

According to (12) and (24) for every half a period ($\omega t=\pi m$, where m is a positive integer) we have

$$\Delta b_1^2=\tfrac{1}{4}-g^2\omega^2/(4g^2\omega^2+2u^2)\,,$$
$$\Delta b_2^2=\tfrac{1}{4}+g^2\omega^2/2u^2,\quad \Delta b_1\Delta b_2=\tfrac{1}{4}\,, \tag{25}$$

while at moments $t=\pi(m+\frac{1}{2})$, Δb_1^2 and Δb_2^2 exchange their values. Just after the short-pulse excitation the state $|u\rangle$ shows a considerably narrower spatial distribution than the vacuum state, while after a quarter of the vibrational period, on the contrary, its spatial distribution becomes wider than that of the vacuum state. The squeezing is especially appreciable if $g\gg u/\omega\gg 1$. In this case $\Delta b_1^2(0)\simeq u^2/8g^2\omega^2$.

Finally, it is worth noting that the vibrational state $|u,t\rangle$ leads to temporal modulation of the spectral characteristics of various optical processes connected with secondary transitions from the excited electronic state of the molecule. In particular, in the time behavior of the spontaneous emission spectrum for the reverse electronic transition $j\rightarrow i$, along with earlier predicted oscillations of the Stokes shift,[14] an additional modulation of the linewidth takes place due to the oscillations of Δb_1. Squeezing of Δb_1 up to $u/2\sqrt{2}g\omega$ causes narrowing of the linewidth of the time-resolved spontaneous emission spectrum up to u, i.e., to the spectral width of the exciting pulse. It is not improbable that such types of spectrum width modulations have already been observed in pump-probe spectroscopical experiments in dye solutions.[15] Time-dependent coordinate fluctuations of the excited molecule might also be important for two-step selective photochemical processes proposed in Refs. 16 and 17.

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