Quantum State Holography

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We introduce a method for reconstructing quantum superposition states excited in molecules or atoms by short laser pulses. The technique is based on mixing the unknown *object state* with a known *reference state* generated in the same system by an additional delayed laser pulse, and detecting the total time- and frequency-integrated fluorescence as a function of the delay. We demonstrate the feasibility of the method by reconstructing various vibrational wave packets in sodium dimers. [S0031-9007(98)05330-7]

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The problem of how to measure the wave function of a quantum system in amplitude *and* phase has attracted a lot of attention over the last few years [1]. Such a reconstruction has been achieved for only a few systems, such as a single mode of the electromagnetic field [2,3], vibratory states of molecules [4], a single ion stored in a Paul trap [5], and an atomic beam [6]. In this Letter, we address the problem of measuring the quantum state of a molecule [4,7,8], and propose the concept of *quantum state holography*.

In analogy to the ordinary optical holography, we interfere the *object* wave function to be measured with a known *reference* wave function. For this purpose, we employ a sequence of two time-delayed laser pulses which subsequently excite the object and the reference wave packet in the excited molecular potential. This is similar to the recently introduced technique of wave packet cross-interferometry [9]. The total incoherent fluorescence [10] of the excited molecule, which is recorded as the function of the delay time τ, serves as a hologram. We show that these data contain enough information to extract the full quantum state of the object wave packet. Moreover, we demonstrate the feasibility of quantum state holography by numerically simulating the reconstruction of wave packets in a sodium dimer. The method is capable of determining wave packets in arbitrary potentials and is not restricted to weakly anharmonic molecules.

Quantum state holography applied to a wave packet in an excited molecular potential adds coherently the *reference* state $|\psi_r\rangle = e^{i\phi(\tau)}\sum_n b_n|n\rangle$ to the *object* state $|\psi_o\rangle = \sum_n a_n e^{-i\omega_n\tau}|n\rangle$ which already has evolved for a time τ. Here, $|n\rangle$ denotes the nth vibrational state with energy $E_n = \hbar\omega_n$ in the excited molecular potential, and we allow for an additional phase $\phi(\tau)$ between the object and reference states. This relative phase is determined by the actual experimental setup and may depend on the delay time τ. In the weak field limit, the population P_n of the nth vibrational level is thus given by

$$P_n(\tau) = |a_n|^2 + |b_n|^2 + 2\,\mathrm{Re}\{a_n b_n^* e^{-i[\omega_n\tau+\phi(\tau)]}\}. \quad (1)$$

Our technique relies on the observation that the interference term in Eq. (1) depends on the *amplitudes* a_n and b_n, rather than on the probabilities $|a_n|^2$ and $|b_n|^2$. As in ordinary holography, the population P_n plays the role of the photographic density, which allows for the full reconstruction of the object wave function. We also note that there is no need to determine the population P_n for each of the states separately: It suffices to measure the time- and frequency-integrated fluorescence of the molecule making a spontaneous transition to the lower electronic level as a function of the delay time τ. Indeed, for a wave packet which comprises several vibrational states $|n\rangle$ populated with the probability P_n, the time-integrated energy emitted incoherently by the molecule is given by

$$F_{\mathrm{tot}} \propto \sum_{\nu,n} P_n|\langle\nu\mid n\rangle|^2 E_{n,\nu}^4\,, \quad (2)$$

where ν numerates vibrational levels in the lower electronic state, and $E_{n,\nu} = E_n - E_\nu$ denotes the energy difference of the two states. From Eqs. (1) and (2) we recognize that in this case the total fluorescence

$$F_{\mathrm{tot}}(\tau) = F_o + F_r + F_{\mathrm{int}}(\tau)\,, \quad (3)$$

consists of the τ-independent terms $F_o \equiv \sum_n |\tilde{a}_n|^2$ and $F_r \equiv \sum_n |\tilde{b}_n|^2$, and the interference term

$$F_{\mathrm{int}}(\tau) \equiv 2\,\mathrm{Re}\left\{\sum_n \tilde{b}_n^*\tilde{a}_n e^{-i[\omega_n\tau+\phi(\tau)]}\right\}, \quad (4)$$

which results from the overlap between the two wave packets, and, therefore, depends on τ. Here, we have introduced the notation $\tilde{a}_n \equiv \kappa_n a_n$ and $\tilde{b}_n \equiv \kappa_n b_n$ with $\kappa_n \equiv [\sum_\nu |\langle\nu\mid n\rangle|^2 E_{n,\nu}^4]^{1/2}$. Note that by measuring the fluorescence of the object and the reference wave packet alone, one finds the quantities F_o and F_r which can be subtracted from F_{tot}. We, therefore, in the remainder concentrate on F_{int} only.

We assume that the reference state $|\psi_r\rangle$ and the coefficients b_n are known quantities. Hence, the only

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unknowns which enter the expression Eq. (4) for the fluorescence $F_{\text{int}}(\tau)$ are the amplitudes a_n of the unknown object wave packet. But how can we recover these complex-valued amplitudes from the real-valued signal $F_{\text{int}}(\tau)$?

In wave packet interferometry one uses a Michelson interferometer with a delay line to prepare the sequence of two laser pulses. During the coherent interaction of the pulses with the molecule the relative phase $\phi(\tau) \equiv \phi_0 + \omega_L \tau$ between the two pulses is transferred onto the wave packets. Here, we allow for an additional phase shift ϕ_0 as in the experiments reported in Ref. [11]. We now measure the fluorescence $F_{\text{int}}(\tau)$ at a fixed delay τ for two different phase angles ϕ_0, e.g., $\phi_0 = 0$ and $\phi_0 = -\pi/2$, and define [12] the signal

$$S(\tau) \equiv \frac{1}{2}\{F_{\text{int}}(\tau, \phi_0 = 0) + iF_{\text{int}}(\tau, \phi_0 = -\pi/2)\} = \sum_n \tilde{b}_n^* \tilde{a}_n e^{-i\{\omega_n + \omega_L\}\tau}. \tag{5}$$

The phase proportional to the optical laser frequency ω_L gives rise to fast oscillations in the signal, the so-called "Ramsey fringes," which can be resolved with current techniques [11,13].

Since every physical state characterized by the coefficients a_n is normalized, these coefficients vanish for a sufficiently large index n. Therefore we assume that only a finite number of coefficients a_n, say $n = 0, \ldots, n_{\max}$, have nonvanishing values. We now make at least $N = n_{\max} + 1$ measurements of the signal $S(\tau)$ at distinct times τ_m with $m = 1, \ldots, N$, and write the resulting set of equations in matrix form,

$$S(\tau_m) = \sum_{n=0}^{n_{\max}} e_{m,n} \tilde{a}_n \,, \tag{6}$$

where we have introduced the notation $e_{m,n} \equiv \tilde{b}_n^* \exp[-i(\omega_n + \omega_L)\tau_m]$.

By numerically inverting the set of equations (6) we can extract the quantities $\tilde{a}_n$ and hence reconstruct the amplitudes a_n of the object wave packet in their modula *and* phases. For the inversion of the set of equations (6) we have used the method of "singular value decomposition" [14]. This method allows us to invert quadratic as well as nonquadratic matrices (corresponding to an overdetermined set of equations). In the latter case it produces a solution that is the best approximation in the least-squares sense. We emphasize that our reconstruction scheme is numerically robust, since the unknown coefficients a_n enter the set of equations (6) only linearly.

We now demonstrate the feasibility of quantum state holography using the two Morse-like potential curves [15] $X(^1\Sigma_g^+)$ and $A(^1\Sigma_u^+)$ of the sodium dimer, for which we calculate numerically the corresponding vibrational eigenstates and energies using the renormalized Numerov method [16]. As the object wave function we choose a molecular Schrödinger cat state [17,18] prepared in the upper electronic potential. Its Wigner function

$$W(r,p) = \frac{1}{\pi\hbar}\int_{-\infty}^{\infty} dy\, \psi^*(r-y)\psi(r+y)e^{-\frac{i}{\hbar}2yp} \tag{7}$$

in r-p phase space is shown in Fig. 1(a), and consists of two distinct wave packets with a prominent fringe structure reflecting the quantum coherence between them. Such a state can be prepared by a pair of delayed laser pulses which pump the system from the ground vibrational level $|\nu = 0\rangle$ [19]. We note that similar states were created only recently for an electronic wave packet in a Rydberg atom [11], for a single atom stored in a Paul trap [20], and for entangled system "atom + electromagnetic field" in a cavity [21]. As the reference state we use a replica of the ground vibrational state in the lower potential, which may be transferred to the upper potential by a short laser pulse. In the limit of short reference pulses, the amplitudes b_n of the excited reference wave packet read simply $b_n = \alpha\langle n \,|\, \nu = 0\rangle$, where α is a constant.

Figure 2 presents the fluorescence hologram for the interval from $\tau = 0$ to $\tau = 2T_{\text{vib}}$. The signal consists of fast oscillations with the optical laser frequency ω_L that are modulated by an envelope which varies with a period of about $T_{\text{vib}}/2$. In order to simulate errors in an actual measurement and to test the robustness of the reconstruction procedure, we have introduced random fluctuations in the measured signal, which obey a Gaussian distribution with standard deviation σ. Here, we have chosen the value $\sigma = 0.1(F_0 + F_r)$ and have taken $N = 100$ simulated data points equally spaced between $\tau = 0$ and $\tau = 2T_{\text{vib}}$.

Figure 1(b) shows the Wigner function of the so reconstructed superposition state. A comparison with Fig. 1(a) demonstrates that the method reproduces nicely the object state. As a measure for the fidelity of the reconstruction procedure we use the overlap $o \equiv |\sum_n a_n^* a_n^{(\text{rec})}|$ between the exact and the reconstructed state, which in this case is $o = 0.98$.

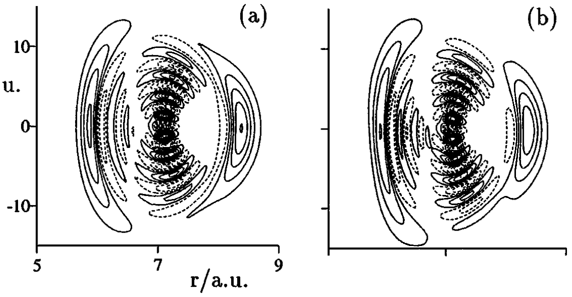

FIG. 1. Quantum state holography: Exact (a) versus reconstructed (b) Wigner function of a molecular Schrödinger cat state. Here, solid lines correspond to positive values of the Wigner function whereas dashed lines correspond to negative values.

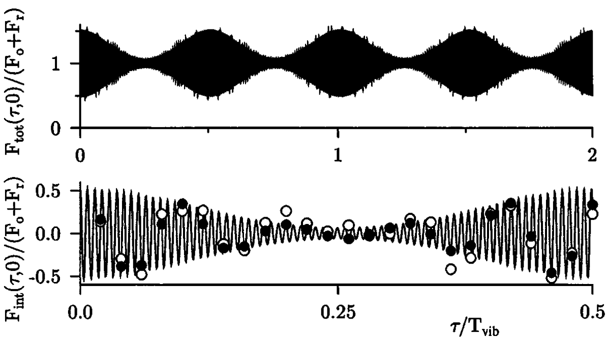

FIG. 2. Fluorescence hologram—a record of the emitted energy of the molecule as a function of the delay τ between the object and reference pulse. In the top we show the complete time region used for the reconstruction, whereas in the bottom we display only a small window around $\tau = 0.25T_{\mathrm{vib}}$. Here, white circles correspond to the simulated data points including measurement errors, whereas black circles correspond to the exact values.

We now discuss the role of the reference state in the performance of quantum state holography. For this purpose we reconstruct the squeezed and rotated state [22]

$$\psi(r) = (\pi u)^{-1/4} e^{ip_0 \frac{(r - r_0/2)}{\hbar}} \exp\left[-(1 - iv)\frac{(r - r_0)^2}{2u} \right] \tag{8}$$

whose Wigner function is shown in Fig. 3(a). Such a state can be created by different methods, as discussed, for example, in Ref. [23].

In Fig. 3(b) we display the Wigner function of the reconstructed state when the ground state $|\nu = 0\rangle$ of the lower potential serves as a reference state. A comparison with the exact Wigner function shows that in this case the reconstruction was only partially successful. Although the main features of the state could be recovered, the wings of the reconstructed Wigner distribution differ from the exact ones. This indicates that coefficients a_n with large values of n were discriminated in the reconstruction. Indeed, from Eq. (4) we find that only those states contribute to the interference fluorescence signal for which the amplitudes *both* of the signal state a_n *and* of the reference state b_n are nonzero. Hence, in order to reconstruct all contributing amplitudes a_n which build up the quantum state to be measured, the reference wave packet has to cover essentially the same energy range as the object state. Figure 4 shows the probabilities $P_n = |a_n|^2$ for the squeezed state Eq. (8) together with the probabilities $P_n = |b_n|^2$ for the two different reference states, $|\nu = 0\rangle$ and $|\nu = 4\rangle$. Whereas $|\nu = 0\rangle$ does not cover the complete energy region of the squeezed state, the state $|\nu = 4\rangle$ does. Hence for the reconstruction of the squeezed state Eq. (8) the reference state $|\nu = 4\rangle$ seems to be more suited. In Fig. 3(c) we show the Wigner function of the squeezed state obtained with the help of this reference state. Again measurement errors were included in the simulation as before. In this case, however, we find a better agreement between the exact and reconstructed Wigner functions, as expressed by the fidelity parameter $o = 0.94$ in contrast to the value $o = 0.87$ obtained with the reference state $|\nu = 0\rangle$.

To conclude, we have introduced the method of quantum state holography to reconstruct the complete state of molecular wave packets in excited electronic potentials. In contrast to emission tomography [4], our technique works for arbitrary potentials, and recovers even highly nonclassical features of quantum states. Compared to the

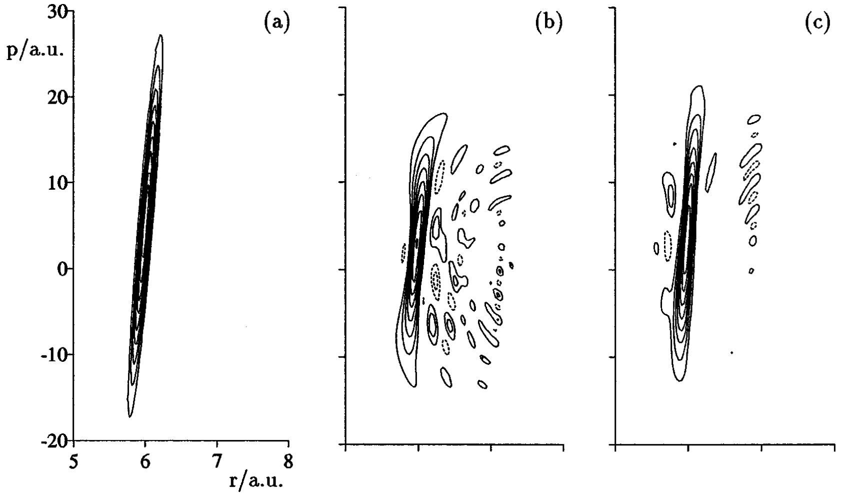

FIG. 3. Role of the reference state in the performance of quantum state holography. (a) shows the contour lines of the exact Wigner function of a squeezed state, whereas (b) and (c) depict reconstructed states. For the reconstruction in (b) we have used the ground state $|\nu = 0\rangle$ and in (c) the state $|\nu = 4\rangle$. We have incorporated measurement errors obeying a Gaussian distribution with variance $\sigma = 0.1(F_o + F_r)$ and have chosen $N = 200$ simulated data points equally spaced between $\tau = 0$ and $\tau = 600$ fsec. The parameters of the squeezed state are $u = 0.025$ a.u., $p_0 = 5$ a.u., $r_0 = 6$ a.u., and $v = 2.1$.

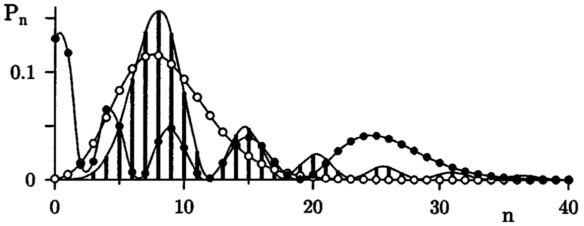

FIG. 4. Energy level population $P_n = |a_n|^2$ (bars) for the object state, that is, the squeezed state of Fig. 3(a), compared to the populations $|\langle n \mid \nu\rangle|^2$ in the reference states $|\nu = 0\rangle$ (white circles) and $|\nu = 4\rangle$ (black circles). To guide the eye we have connected the discrete values by continuous curves.

recently introduced method of "wave function imaging" [7], the quantum state holography does not require time-resolved fluorescence detection. In addition, our linear reconstruction technique is much more numerically robust than the nonlinear procedure of Ref. [7]. Although the specific wave packets considered in this Letter belong to the discrete part of spectrum, our preliminary results indicate that after proper modification the procedure may be applied to the dissociating states as well. Moreover, this method may be extended to reconstruct the quantum state of an electronic wave packet in Rydberg atoms. We also note that quantum state holography might develop into a powerful tool to determine the spectral amplitudes and phases of ultrashort laser pulses: Here we use the molecule as a "grating" to chop the unknown pulse into its spectral amplitudes and phases which we later read out with the help of a simple reference pulse.

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[1] For a review, see the special issue on quantum state preparation and measurement, edited by W. P. Schleich and M. Raymer [J. Mod. Opt. **44**, No. 11/12 (1997)]; M. Freyberger *et al.,* Phys. World **10**, 11 (1997).

[2] N. G. Walker and J. E. Carrol, Opt. Quantum Electron. **18**, 355 (1986); J. W. Noh, A. Fougeres, and L. Mandel, Phys. Rev. Lett. **67**, 1426 (1991).

[3] D. T. Smithey *et al.,* Phys. Rev. Lett. **70**, 1244 (1993); S. Schiller *et al.,* Phys. Rev. Lett. **77**, 2933 (1996); G. Breitenbach *et al.,* Nature (London) **387**, 471 (1997).

[4] T. J. Dunn *et al.,* Phys. Rev. Lett. **74**, 884 (1995).

[5] D. Leibfried *et al.,* Phys. Rev. Lett. **77**, 4281 (1996).

[6] Ch. Kurtsiefer *et al.,* Nature (London) **386**, 150 (1997).

[7] M. Shapiro, J. Chem. Phys. **103**, 1748 (1995); Chem. Phys. Lett. **242**, 548 (1995); Chem. Phys. **207**, 317 (1996).

[8] U. Leonhardt and M. G. Raymer, Phys. Rev. Lett. **76**, 1985 (1996). In this paper the quantum state of a one-dimensional wave packet is inferred from its time-dependent position distributions.

[9] J. F. Christians and B. Broers, Phys. Rev. A **52**, 3655 (1995); D. W. Schumacher *et al.,* Phys. Rev. A **52**, 4719 (1995).

[10] In contrast to Refs. [4,7] we use here the time- and frequency-*integrated* fluorescence as the measured signal.

[11] M. W. Noel and C. R. Stroud, Jr., Phys. Rev. Lett. **75**, 1252 (1995); **77**, 1913 (1996).

[12] This is similar to the method of endoscopy proposed for a cavity field in P. J. Bardroff *et al.,* Phys. Rev. A **51**, 4963 (1995), and for an ion in a trap by P. J. Bardroff *et al.,* Phys. Rev. Lett. **77**, 2198 (1996).

[13] V. Blanchet *et al.,* Chem. Phys. Lett. **233**, 491 (1995).

[14] W. H. Press *et al., Numerical Recipes* (FORTRAN version) (Cambridge University Press, Cambridge, England, 1989).

[15] I. Schmidt, Ph.D. thesis, Universität Kaiserslautern, 1987.

[16] B. R. Johnson, J. Chem. Phys. **67**, 4086 (1977).

[17] J. Janszky *et al.,* Phys. Rev. A **50**, 1777 (1994).

[18] I. A. Walmsley and M. G. Raymer, Phys. Rev. A **52**, 681 (1995).

[19] The coefficients of the so prepared state read

$$a_n = \mathcal{N}\langle n \mid \nu = 0\rangle \exp\left(-\frac{\Delta_n^2\Delta_t^2}{2}\right)\{1 + e^{-i\Delta_n\delta t}\},$$

where $\mathcal{N}$ is a normalization constant, and $\Delta_n \equiv \omega_n - \omega_g - \omega_L$ denotes the detuning for the nth vibrational level. The carrier frequency ω_L of the laser is chosen to be resonant with the absorption maximum close to the $n = 8$ level. For the numerical values of Δ_t and δt we use $\Delta_t = 0.1T_{\text{vib}}$ and $\delta t = 0.5T_{\text{vib}}$, where the vibrational period equals $T_{\text{vib}} = 300$ fsec for the specified group of levels.

[20] C. Monroe *et al.,* Science **272**, 1131 (1996).

[21] M. Brune *et al.,* Phys. Rev. Lett. **77**, 4887 (1996).

[22] B. Dutta *et al.,* J. Opt. Soc. Am. B **10**, 253 (1993).

[23] D. G. Abrashkevich *et al.,* J. Chem. Phys. **101**, 9295 (1994).