Coherent Propagation and Quantum Beats of Quadrupole Polaritons in Cu_2O

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(Received 1 July 1991)

Coherent propagation of quadrupole polaritons in Cu_2O is demonstrated by time-resolved spectroscopy. This manifests itself in a strong distortion of the temporal shape of a picosecond optical pulse which is in resonance with the $1S$ exciton polariton. Oscillations with time-dependent period in the transmitted light intensity are quantitatively explained as resulting from quantum beats between the two branches of the exciton polariton. The analysis yields the homogeneous linewidth and oscillator strength.

PACS numbers: 71.36.+c, 42.50.Md, 78.47.+p

Coherent optical spectroscopy is a very powerful technique to investigate the dynamics of nonstationary states. For instance, the observation of quantum beats yields very direct information about the coherence properties of electronic or vibronic excitations, as was shown for atoms and molecules in many experiments [1]. Recently, several groups have succeeded in observing quantum beats of extended states such as excitons in semiconductors, where the much faster dephasing processes require ultrafast laser and detection techniques [2–5]. In all experiments so far the decay of exciton coherence was observed in physical situations where propagation effects were of no relevance.

In this Letter we report the first observation of corresponding coherent phenomena from polaritons. The results are novel for two reasons: First, in contrast to the previous cases of quantum beats, only one exciton state is needed. The coupling of the exciton with light automatically leads to two branches which can interfere. Second, polaritons are propagating modes. As a consequence, the observation of quantum beats is the signature of conservation of coherence over macroscopic distances.

The problem of pulse propagation in a dispersive medium has been addressed by several authors [6–8]. Puri and Birman [6] discuss different situations which are characterized by the ratio of polariton damping constant Γ and spectral width τ^{-1} of the incoming Gaussian pulse. In our case, we have the interesting situation of $\Gamma\tau \ll 1$. The authors predict a broadening and a distortion of the pulse and even oscillations. In our experiments we measure the time-resolved transmission of an optical pulse, which is tuned to the $1S$ exciton resonance in Cu_2O. Indeed, we observe a strong distortion of the pulse shape as well as pronounced oscillations. Surprisingly, the period of these oscillations depends on the delay time and the thickness of the crystal. The oscillations are interpreted as quantum interference between polaritons of the two very closely spaced branches of the quadrupole resonance.

For several reasons the quadrupole polariton of the lowest $1S$ exciton transition of Cu_2O is ideally suited for the study of coherent propagation effects: (i) The oscillator strength of the quadrupole transition is very small ($f \sim 10^{-9}$, Ref. [9]). This leads to a small splitting of the polariton into two branches, and, therefore, to a narrow region of strong dispersion in (ω,k) space. (ii) There exist samples of high structural quality so that scattering from static imperfections can be neglected. (iii) Exciton–acoustic-phonon scattering, which is the major remaining cause of polariton dephasing, is strongly reduced at low temperatures ($T \leq 2$ K) due to the small number of available final states. (iv) Although Cu_2O is cubic (O_h symmetry), the oscillator strength of the quadrupole absorption is anisotropic. By proper choice of the k vector and polarization direction one can thus excite polariton branches with different splitting. We therefore expect a dependence of the beat period on the excitation conditions (k vector and polarization direction).

For the observation of polariton propagation we employ two setups. Both allow measurements with high time and spectral resolution. In the first setup we use the second harmonic of an active-mode-locked and Q-switched Nd-doped yttrium-aluminum-garnet (YAlG) laser for the synchronous pumping of two dye lasers with a repetition rate of 10 Hz. One dye laser beam (pulse energy up to 0.1 mJ, pulsewidth of 60 ps, and spectral width of 70 μeV) is sent through the sample, whereas the other beam is sent as a time reference through a variable delay. Both beams are then focused on a KDP crystal for sum frequency generation. The intensity of the sum frequency signal is measured with a monochromator and a photomultiplier as a function of the delay between the two dye laser pulses. With the use of two dye lasers, which are tuned to different photon energies, we avoid problems

due to second-harmonic generation for each laser beam, which would lead to background light independent of the delay between the pulses. In some measurements we use pulses of the YAlG laser as the time reference instead of the second dye laser. With this setup we are able to measure signals with delays up to 6 ns with a background suppression of 6 orders of magnitude. The intensity on the sample can be increased to well above 10^9 W/cm^2 by proper focusing.

In the second setup we use a mode-locked Nd-doped yttrium-lithium-fluoride laser, which is frequency doubled to synchronously pump a dye laser with a repetition rate of 76 MHz (average power of 10 mW, pulsewidth of 30 ps, and spectral width of 25 μeV). The measurement of the time profile of the pulses is achieved by time-resolved photon counting. We use a microchannel-plate photomultiplier behind a double monochromator with subtractive dispersion in order to avoid any transit time spread. The overall time resolution is 60 ps (FWHM). For details of this optical setup and the electronics we refer to recent publications [10].

For the measurements we use high-quality natural crystals of Cu_2O which were oriented by x-ray diffraction. The crystals are cooled down to about 1.8 K in a helium cryostat.

Figure 1 shows an experimental result obtained with the second setup for a crystal of thickness $d=0.91$ mm and light propagation along a [110] direction. The laser was tuned to the $1S$ exciton at $\hbar\omega_0=2.0329$ eV and was polarized along [001], since the quadrupole transition is allowed for this configuration. Note that the transmitted pulse has acquired a long-lasting trailing edge. Superimposed on this long decay we find a clearly resolved modulation, the period of which increases by more than a factor of 5 within the first 2.5 ns. If it were not for this time-dependent period, the decay would resemble a typical beat signal of two closely lying states, which are coherently excited. We are able to explain the striking time dependence of the oscillation period quantitatively by an analysis of the polariton propagation.

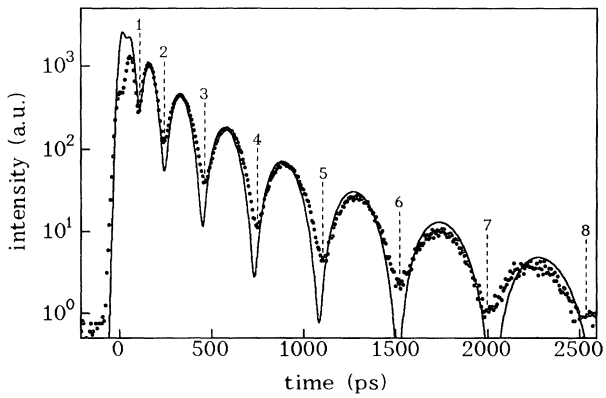

FIG. 1. Temporal evolution of a 30-ps laser pulse tuned to the $1S$ exciton resonance in a Cu_2O crystal of thickness $d=0.91$ mm. k vector and polarization direction are along [110] and [001], respectively. Solid circles denote experimental results; the solid line denotes the numerical calculation. The numbers mark the values of n, in accordance with Eq. (4).

The propagation properties of polaritons are determined by the dielectric function $\varepsilon(\omega,k)$. In the region of a quadrupole it is given by [11]

$$\varepsilon(k,\omega)=\left(\frac{ck}{\omega}\right)^2=\varepsilon_b+\frac{fc^2k^2}{\omega_0^2+ak^2-\omega^2-i\omega\Gamma}, \quad (1)$$

where $\varepsilon_b=6.5$ is the background dielectric constant, $\hbar\omega_0=2.0329$ eV is the $1S$ exciton energy, and $ak^2=\omega_0\hbar k^2/M$ accounts for spatial dispersion with $M=m_e+m_h=2.7m_0$ for the total exciton mass. Γ is a phenomenological damping rate that accounts for the dephasing of the states and $f=f_{hkl}$ is the oscillator strength, where $[hkl]$ is the direction of the k vector. Equation (1) differs from the usual dielectric function for a dipole transition by the k^2 factor in the numerator. Also, the quadrupole oscillator strength is smaller by at least 5 orders of magnitude as compared to a dipole-allowed transition. This results in a very small splitting of the polariton branches, as seen in Fig. 2(a).

For a quantitative fit of the experimental results we have to start with the Fourier components of the polariton pulse at the entrance of the crystal which propagate according to the dispersion of the quadrupole resonance to the rear side of the crystal. We then calculate the electric field $E_s(z,t)$ of the signal beam as a function of time by the Fourier integral of its frequency distribution

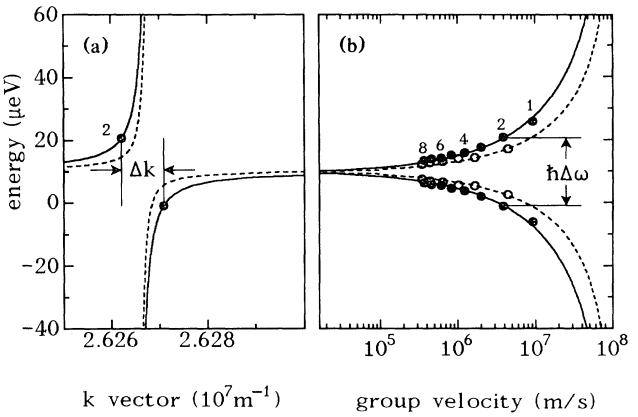

FIG. 2. (a) Calculated dispersion of $1S$ exciton polariton in Cu_2O, and (b) the corresponding group velocity. The zero energy scale refers to $\hbar\omega_0=2.0329$ eV. Solid lines denote oscillator strength $f_{110}=3.6\times10^{-9}$; dashed lines denote $f_{111}=1.4\times10^{-9}$; solid circles refer to the experimental time-dependent beat period of Fig. 1, numbered by their values of n; open circles refer to experimental results for the k vector along a [111] direction.

at $z=d$:

$$E_s(z,t) \propto \int_{-\infty}^{\infty} \sum_{j=1}^{2} a_j(k_j,\omega) E_0(\omega) \exp[i(k_j z - \omega t)] d\omega . \quad (2)$$

The sum over j accounts for the two polariton branches which are excited with amplitudes $a_j(k_j,\omega)$. These amplitudes are calculated with the use of Pekar's additional boundary conditions [12]. $E_0(\omega)$ is the Fourier amplitude of the exciting laser pulse; its amplitude profile is taken as a Lorentzian centered at $\hbar\omega_0$ with FWHM of 25 μeV. The detected signal $S(t)$ is calculated by convoluting the signal intensity with the response function $Z(t)$ of the detection system. $Z(t)$ is approximated by a Gaussian function with FWHM of 40 ps. So,

$$S(t) \propto \int_{-\infty}^{\infty} |E_s(t-\tau)|^2 Z(\tau) d\tau . \quad (3)$$

The result of the numerical calculation is shown as a solid line in Fig. 1. The oscillator strength f and the damping constant Γ are taken as adjustable parameters for a best fit. The period of oscillations is very sensitive to the value of f, which can thus be determined with high accuracy. We find $f_{110}=3.6\times10^{-9}$ in good agreement with a value reported in Ref. [9] and a value of $\Gamma=6.5\times10^{-7}$ eV. We have also fitted experimental curves obtained on different crystal thicknesses by the use of the same parameters f and Γ and the appropriate value of d. As expected we observe that at a given delay the period decreases with increasing thickness.

As mentioned before, the quadrupole oscillator strength depends upon the direction of light propagation and the polarization orientation. For instance, if the wave vector is chosen along a [111] direction, one expects a reduced oscillator strength $f_{111}=\frac{1}{3}f_{100}=\frac{1}{3}f_{110}$ with a corresponding dispersion curve shown as dashed lines in Fig. 2. From our experiments in the [111] configuration we derive $f_{111}=1.4\times10^{-9}$, which is close to $\frac{1}{3}f_{110}=1.2\times10^{-9}$.

A more intuitive explanation for the appearance of the oscillations and of their time- and thickness-dependent period is given as follows. Consider for simplicity a pair of pulses, one from each branch, propagating with the same group velocity v_g. During propagation along z the pulses experience a relative phase shift $\Delta\Phi(z)$, which depends both on the difference $\Delta\omega$ in frequency and Δk in wave vector of the polariton wave packets,

$$\Delta\Phi(z) = \Delta k z - \Delta\omega z / v_g . \quad (4)$$

Superposition of these pulses then results in a time-dependent total intensity. Clearly, in order to show beats both pulses must experience little damping and dephasing as they propagate through the sample. The time dependence of the beat period can be understood by considering the group-velocity dependence of the energy splitting $\hbar\Delta\omega$ and the difference in wave vector Δk which follows from Eq. (1). In Fig. 2(b) we have translated the time-varying beat period of Fig. 1 into an energy splitting as a function of group velocity (solid circles). For the analysis we consider a minimum at the time $t=d/v_g$. For this minimum the phase difference [Eq. (4)] at the end of the crystal has to fulfill the condition $\Delta\Phi(d)=(2n+1)\pi$, with $n=0,1,\ldots$ numbering the subsequent minima. For these minima we thus get discrete values of the corresponding group velocity, although the group velocity varies continuously. In Fig. 2(b) we could have plotted the maxima or points for any phase in between [see Eq. (4)] which would then cover the whole range of group velocities. Since the splitting decreases for lower group velocities we expect a corresponding increase of the beat period at later times. The experimental results confirm this interpretation. We have marked the minima by their values of n in Figs. 1 and 2(b).

We have performed the same analysis for propagation along a [111] direction, where the oscillator strength is reduced by a factor of 3 (dashed lines in Fig. 2). The open circles mark the minima, which are gained from the experimental results with the use of Eq. (4). By the same token, for a thicker sample a given delay corresponds to a larger group velocity, resulting in a shorter period, which is confirmed by experiments. This simple picture also explains why polariton beats can only occur if both polariton branches are simultaneously excited. We have verified this point experimentally by slightly detuning the laser from the resonance frequency.

For the measurements of Fig. 1 the intensity was kept well below 10^2 W/cm^2. With the use of the first setup, we performed experiments with intensities up to 4×10^9 W/cm^2. Intensities higher than 10^8 W/cm^2 lead to a faster decay and a clear reduction of the beat amplitude. We believe that this is due to exciton-exciton interaction near the front surface resulting in a loss of coherence. We also observed a similar trend by raising the temperature, which is probably due to scattering by acoustic phonons. Further detailed measurements of the temperature dependence are necessary for a quantitative investigation of this point.

With an external magnetic field the polariton can be split. This should lead to additional beats due to the coherent excitation of the split states. We have indeed observed pronounced oscillations in magnetic fields up to 0.5 T, the frequency of which scales with the applied field.

In conclusion, we report the first observation of polariton beats. This is clear evidence for coherent propagation of the quadrupole excitation over macroscopic distances. The observed oscillations and the change of their period with time is explained by quantum interference between the excitations of the two polariton branches. The analysis yields a spectacular small homogeneous linewidth of $\Gamma=6.5\times10^{-7}$ eV corresponding to a coherence time $\tau_{coh}=2\hbar/\Gamma=2$ ns and a precise value for the oscilla-

tor strength ($f_{110}=3.6\times10^{-9}$).

We thank G. Nattkemper for help with the experiments. We acknowledge the financial support of this project by the Deutsche Forschungsgemeinschaft and a DFG-CNRS travel support.

[1] S. Haroche, in *High Resolution Laser Spectroscopy,* edited by K. Shimoda (Springer-Verlag, Heidelberg, 1976).

[2] V. Langer, H. Stolz, and W. von der Osten, Phys. Rev. Lett. **64**, 854 (1990).

[3] E. O. Göbel, K. Leo, T. C. Damen, J. Shah, S. Schmitt-Rink, W. Schäfer, J. F. Müller, and K. Köhler, Phys. Rev. Lett. **64**, 1801 (1990); K. Leo, T. C. Damen, J. Shah, E. O. Göbel, and K. Köhler, Appl. Phys. Lett. **57**, 19 (1990).

[4] B. F. Feuerbacher, J. Kuhl, R. Eccleston, and K. Ploog, Solid State Commun. **74**, 1279 (1990).

[5] S. Bar-Ad and I. Bar-Joseph, Phys. Rev. Lett. **66**, 2491 (1991).

[6] A. Puri and J. L. Birman, Phys. Rev. A **27**, 1044 (1983); in *Semiconductors Probed by Ultrafast Spectroscopy II,* edited by R. R. Alfano (Academic, New York, 1984), pp. 295–330.

[7] D. L. Johnson, Phys. Rev. Lett. **41**, 417 (1978).

[8] J. A. Gaspar-Armenta and P. Halevi, Opt. Commun. **64**, 217 (1987).

[9] S. Nikitine, J. B. Grun, and M. Certier, Phys. Kondens. Mater. **1**, 214 (1963); P. D. Bloch, B. Meyer, and C. Schwab, J. Phys. C **13**, 267 (1980).

[10] H. Stolz, in *Festkörperprobleme: Advances in Solid State Physics,* edited by U. Rössler (Vieweg, Braunschweig, 1991), Vol. 31; V. Langer, H. Stolz, and W. von der Osten, J. Lumin. **45**, 406 (1990).

[11] V. M. Agranovich and V. L. Ginzburg, in *Crystal Optics with Spatial Dispersion and Excitons,* edited by V. M. Agranovich and V. Ginzburg, Springer Series in Solid State Sciences Vol. 42 (Springer-Verlag, Heidelberg, 1984).

[12] S. I. Pekar, V. N. Piskovoi, and B. E. Tsekvava, Fiz. Tverd. Tela (Leningrad) **23**, 1905 (1981) [Sov. Phys. Solid State **23**, 1113 (1981)].