Quantum Beats of Excitons in Quantum Wells

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We report the observation of quantum beats in the decay of the coherent polarization of intrinsic excitations in GaAs/GaAlAs quantum wells. The beating arises from interference of excitons with slightly different quantum confinement energy due to well-width fluctuations.

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Time-resolved studies of the loss of coherence of optically excited electronic or vibrational transitions provide one of the most powerful tools to investigate interaction processes of excited states with their environment. The decay of the macroscopic polarization due to dephasing depends not only on scattering processes but also upon the excitation conditions and the nature of the respective transitions, e.g., whether only a single state is excited or several with closely spaced energies. The decay of the polarization is purely exponential, with the dephasing time T_2 as the characteristic time constant, only for the case of a homogeneously broadened transition. For an inhomogeneously broadened transition, the polarization decays faster due to the destructive interference of excitations with slightly different eigenfrequencies. In the particular case of simultaneous coherent excitation of only a few states, e.g., two with slightly different excitation energy, this interference can be resolved as an oscillatory modulation of the polarization decay, which is generally referred to as *quantum beats.*

Interference phenomena[1] and oscillations in the time domain[2] were first observed in the resonance fluorescence of atoms. Later, quantum beats in molecules were observed in fluorescence,[3] in photon-echo signals,[4] in coherent Raman scattering,[5] and recently, in four-wave mixing experiments with femtosecond time resolution.[6] Quantum beats due to the hyperfine splitting in nuclei have also been detected recently in synchrotron-radiation experiments.[7] In solids, spin-echo beats of electronic excitations of F centers[8] and molecular vibrational transitions in organic crystals[9] were reported.

Quantum beats resulting from the interaction of *extended electronic states in solids* have not been reported so far. In fact, the optically coupled continuum states of an intrinsic semiconductor can be considered as an inhomogeneous distribution of two-level systems in momentum space if Coulomb interaction is neglected. The polarization created by an excitation pulse with a given spectral width within this broad inhomogeneous distribution will decay rapidly regardless of scattering. This is because of the destructive interference of all the excited two-level states with different eigenfrequencies, as characteristic of an inhomogeneously broadened transition. Quantum beats therefore are not expected to occur. Including Coulomb interaction, however, makes a fundamental difference, because Coulomb interaction gives rise to the appearance of exciton bound states within the energy gap. Although these states still correspond to a coherent superposition of extended momentum-space states, distributed according to the exciton orbital wave function, their energetic discreteness should make quantum beats observable.

In this Letter, we present the first observation of quantum beats from *extended electronic states in a solid.* In our experiment, we observe quantum beats with terahertz beat frequency in the decay of the coherent exciton polarization in a semiconductor quantum-well (QW) structure. The quantum beat period in the time domain corresponds to the splitting between free-exciton levels observed in luminescence and absorption spectra. The experimental results agree well with a simple theoretical model.

The samples are grown by molecular-beam epitaxy on an n-doped [100] GaAs substrate. The basic sequence consists of a 25-monolayer-thick ($\approx$70-Å) GaAs QW, followed by a 17-monolayer-thick ($\approx$48-Å) $Al_{0.35}Ga_{0.65}As$ barrier, and a 16-monolayer-thick ($\approx$45-Å) GaAs QW.[10] Ten periods separated by 150-Å $Al_{0.35}Ga_{0.65}As$ barriers are grown. Sample B is grown with a growth interruption of 60 s at every interface;

sample A is grown without growth interruptions. The substrate of the samples was removed by etching, thereby allowing transmission experiments. The laser source is a tandem synchronously pumped laser system. A mode-locked YLF laser pumps a rhodamine-6G dye laser, which in turn pumps a LDS750 dye laser, emitting pulses with a second-order intensity autocorrelation width (FWHM) of 800 fs and a spectral width (FWHM) of 4.2 meV tunable between 700 and 800 nm. The sample is mounted in a liquid-He cryostat.

We measure the decay of the phase coherence of resonantly photoexcited excitons by the two-pulse self-diffracted transient-grating technique.[11] Excitation with two perpendicularly polarized pulses creates an excitonic-orientational grating as long as the delay time T between the pulses is of the order of the phase relaxation time. The photons of the second beam will then be diffracted by this transient grating giving rise to a signal into the direction corresponding to $2\mathbf{k}_1-\mathbf{k}_2$, where $\mathbf{k}_1$ and $\mathbf{k}_2$ are the wave vectors of the two incoming beams. The decay of the diffracted signal with increasing delay time between the pulses is therefore a sensitive measure of the phase relaxation time T_2. It has been shown that the decay of the diffracted intensity is exponential, with $T_2/2$ and $T_2/4$ as the characteristic time constant for homogeneously and inhomogeneously broadened systems, respectively.[12]

Figure 1 shows the decay[13] of the diffracted signal as a function of the delay time between the two pulses for an excitation density of about 1×10^9 excitons/cm^{-2} for both samples. Also plotted in the inset of Fig. 1 are the photoluminescence excitation spectra for sample B (lower trace) and sample A (upper trace), which clearly reveal the different absorption spectra of the two samples. Sample A exhibits a broad exciton absorption feature with a spectral width of 3.4 meV (FWHM), whereas in sample B three distinct exciton peaks with a spacing of 2.7 meV and a FWHM of about 1.3 meV can be resolved. The three distinct exciton peaks in sample B correspond to emission of excitons out of regions with different confinement due to thickness differences.[14] In sample A such regions are not present and the inhomogeneous broadening is much stronger.

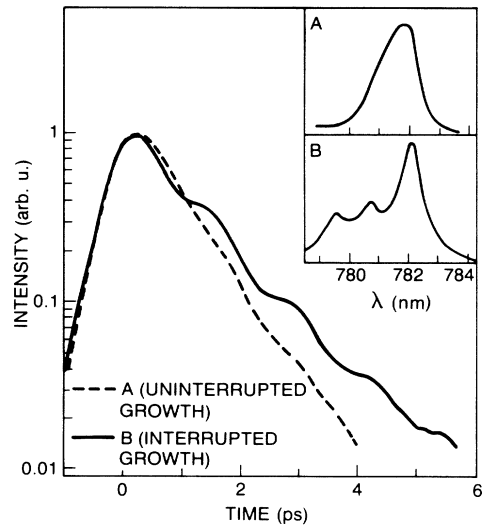

FIG. 1. Decay of the self-diffracted signal as a function of the delay time for sample B (with growth interruption, solid line) and sample A (without growth interruption, dashed line). Excitation density and temperature are about 1×10^9 cm^{-2} and 5 K, respectively. Sample A is resonantly excited at the exciton peak; for sample B, the excitation spectrum covers the two energetically lower exciton levels. Inset: Photoluminescence excitation (PLE) spectra of sample A (top) and sample B (bottom); the exciton splitting in the absorption spectrum of sample B (not shown) agrees with the PLE splitting.

The transient-grating signal is also *fundamentally different for the two samples:* The dashed line shows the result for sample A, which is grown without growth interruption resulting in a broad exciton luminescence line. The decay is nearly exponential with a time constant of about 0.8 ps corresponding to a T_2 of 3.2 ps, assuming an inhomogeneously broadened transition. This result is in reasonable agreement with results reported in Ref. 15.

The coherence decay of *sample B with split exciton levels* (solid line in Fig. 1), however, shows a different behavior: *An oscillatory structure* is superimposed onto the exponential decay. The time constant of the exponential decay of about 1.2 ps is *larger* than in sample A. The period of the superimposed oscillation is about 1.33 ps, corresponding to an energy splitting of 3.1 meV, which is slightly larger than the energy separation of the exciton lines in the photolumininescence excitation (PLE) spectrum. The oscillation frequency does not depend on excitation density or temperature; however, the polarization decay becomes faster with increasing temperature and excitation density. No oscillation is observed in the transient four-wave mixing for the exciton transitions of the narrow well in the same sample, where the splitting of the luminescence lines is larger than the spectral width of our laser pulse; i.e., the exciton states of two thickness regions are not simultaneously excited.

All these observations show that the oscillations observed in the decay of the diffracted signal are *quantum beats caused by the coherent superposition of the polarization of excitons from spatial regions with slightly different energy.* To the best of our knowledge, this is the first observation of quantum beats in the decay of the polarization due to extended state excitations in a solid. In addition, it is the first observation where the energy levels causing the interference are *spatially separated* over large distances.

In order to describe our experimental observations, we make use of the so-called phase-space filling model which has been applied successfully to a number of nonlinear optical phenomena in both inorganic[16] and organic[17] semiconductors. This model ties the nonlinear optical response of excitons to the Fermi statistics of the un-

derlying momentum-space states, resulting in effective anharmonic exciton-photon and exciton-exciton interactions. The latter shall not concern us here; their main effect on two-pulse self-diffraction is the appearance of a signal [$\sim\exp(4T/T_2)$] in a homogeneously broadened system at negative time delays T, as shown recently both experimentally and theoretically.[18]

As has been discussed in the literature,[16,17] for resonant exciton creation (corresponding to our experimental conditions), the third-order anharmonic exciton-photon interaction mediated by phase-space filling is of the same form as that of Frenkel excitons (real-space, two-level systems), but with a modified nonlinear cross section, reflecting the fraction of the Brillouin zone that participates in the exciton wave function. We may therefore follow the pioneering work of Yajima and Taira[12] in modeling our experiments.

We consider a two-peak inhomogeneous exciton distribution of the form[19]

$$\rho(\omega)=\sum_{n=1}^{2}\frac{1}{2\sigma\sqrt{\pi}}\exp\left[\frac{-(\omega-\omega_n)^2}{\sigma^2}\right], \tag{1}$$

where σ is the inhomogeneous broadening. The exciton splitting $\Delta\omega=\omega_1-\omega_2$ is finite for sample B and zero for sample A. In the short-pulse limit, $E_1(\mathbf{r},t)\sim\exp(i\mathbf{k}_1\cdot\mathbf{r})\delta(t-T)$ and $E_2(\mathbf{r},t)\sim\exp(i\mathbf{k}_2\cdot\mathbf{r})\delta(t)$, the time-integrated signal propagating in the direction $2\mathbf{k}_1-\mathbf{k}_2$ is given by

$$I(T)\propto\theta(T)\exp(-4\gamma T)\int_{-T}^{\infty}dt\,[1+\cos(\Delta\omega t)]\times\exp(-2\gamma t-t^2\sigma^2/2), \tag{2}$$

where $\theta(T)$ is the Heaviside step function and $\gamma=T_2^{-1}$ is the homogeneous linewidth. For $\sigma\rightarrow\infty$ (strong inhomogeneous broadening), this expression reduces to

$$\lim_{\sigma\rightarrow\infty}I(T)\propto\theta(T)\exp(-4\gamma T), \tag{3}$$

while for $\sigma\rightarrow 0$ (homogeneous broadening only)

$$\lim_{\sigma\rightarrow 0}I(T)\propto\theta(T)\exp(-2\gamma T)\times\left[1+\frac{\cos(\Delta\omega T)+(\Delta\omega/2\gamma)\sin(\Delta\omega T)}{1+(\Delta\omega/2\gamma)^2}\right]. \tag{4}$$

As discussed above, strong inhomogeneous broadening suppresses quantum beats; however, even for a homogeneously broadened system, the modulation of the signal is never complete.

Figure 2 shows the results of a numerical evaluation of Eq. (2) for parameters listed in the figure caption. The simple theory reproduces the experimental observations remarkably well, except around zero time delay, where the pulse profile comes into play. However, the inhomogeneous broadening of the individual exciton peaks for sample B has to be chosen smaller than the experimental values ($\sigma_{th}=0.2$ meV, $\sigma_{expt}=0.5$ meV). Theoretical curves with $\sigma_{th}=0.5$ meV still show quantum beats with the same period, but the beats are less pronounced compared to the experimental result at large delays. The reason for this difference as well as for the slight discrepancy between the oscillation period in the transient-grating signal and the energy splitting in the PLE spectrum is not fully understood at present. Most important, however, the theory reproduces the quantum beats for sample B whereas they are not predicted for sample A, where the inhomogeneous broadening dominates, in agreement with the experimental findings.

In conclusion, we report the first observation of interference effects (quantum beats) in the coherent emission from extended electronic states in a solid. The occurrence of these beats is a direct consequence of the Coulomb interaction between the laser-excited electron-hole pairs, resulting in a transformation of the optically coupled states from a broad inhomogeneous distribution in k space without Coulomb interaction into an effective two-level system. A simple theoretical model describes the experimental results well. Similar effects should be observable for simultaneous excitation of heavy- and light-hole excitons.

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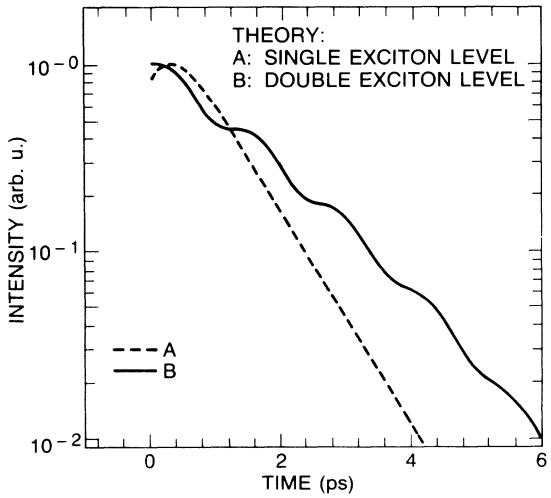

FIG. 2. Calculated decay of the self-diffracted signal as a function of the delay time, according to Eq. (2). The parameters used in the calculation were $\Delta\omega=0$ meV, $\gamma=0.21$ meV, $\sigma=1.80$ meV (sample A, dashed line); $\Delta\omega=3.1$ meV, $\gamma=0.21$ meV, $\sigma=0.20$ meV (sample B, solid line).

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Note added.—After submitting this manuscript, we have indeed observed quantum beats between heavy- and light-hole excitons in a quantum well.

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[19]Because of the spectral width of the excitation laser pulses of 4.2 meV (FWHM) only the two energetically lower of the exciton transitions are excited coherently in the experiments reported here.