Quantum beats of free and bound excitons in GaAs/$Al_xGa_{1-x}As$ quantum wells

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We report the observation of quantum beats due to the interference of the polarization decay of free- and bound-exciton states in a GaAs quantum-well structure.

If two energetically closely spaced transitions are excited with a short optical pulse, a coherent superposition is created. The macroscopic polarization then shows an interference due to the slightly different transition energies; the frequency of this beating is given by $\nu=\Delta E/h$, where $\Delta E=E_2-E_1$ is the difference in the transition energies. These so-called "quantum beats" (QB's) can be observed by techniques like time-resolved fluorescence or wave mixing and are a powerful tool of nonlinear spectroscopy.[1] The interference has been observed for transitions with the same ground state (i.e., three-level systems), which represents an interference of the wave functions, and between separate two-level systems, where the polarization interferes due to the different phase factors of the wave functions.

Recently, QB's originating from *extended* electronic states in solids were observed.[2-7] At a first glance, those observations were surprising: The broad energy bands in a solid can be considered as an inhomogeneous distribution in **k** space. The excitation of such a distribution would lead to a rapid decay of the macroscopic polarization. However, the presence of energetically well-defined *excitonic levels* allows the observation of QB's.[2] We have already reported the first observation of QB's between heavy-hole excitons split by spatially varying confinement[2] and QB's between heavy-hole (hh) and light-hole (lh) excitons.[6] The first system corresponds to independent two-level systems and the second to a three-level system. The latter observation also confirmed the recent observation[8] of a novel coherent polarization interaction.

In this Brief Report, we present the first observation of quantum beats originating from the polarization interference of free heavy-hole exciton states and exciton states bound to an impurity in a semiconductor quantum well. The quantum beats are observed in the decay of the diffracted signal in a self-diffracted degenerate four-wave-mixing experiment. The quantum beat period in the time domain corresponds to the splitting between free- and bound-exciton levels observed in luminescence spectra.

The sample is grown by molecular-beam epitaxy on *n*-type doped [100] GaAs substrate and contains 10 GaAs quantum wells of 170 Å nominal width. The substrate of the sample is removed to allow experiments in a transmission configuration. The sample is mounted in a liquid-helium cryostat and cooled to about 5 K. For the time-resolved experiments, we use a synchronously mode-locked Styryl 8 dye laser, tandem pumped by a rhodamin 6G dye laser and a mode-locked yttrium lithium fluoride laser. The laser pulses have a second-order intensity autocorrelation width (full width at half maximum, FWHM) of 700–800 fs and a spectral width (FWHM) of about 3.5 meV.

Figure 1 shows in the lower part the photoluminescence spectrum (solid line) and the photoluminescence excitation (PLE) spectrum (dashed line) in the vicinity of the first subband heavy-hole (hh) transition, taken at $T_L=2$ K with a low-intensity cw laser. Visible are the hh free-exciton transition at 1.531 eV of about 0.8 meV FWHM and (only weakly visible in the PLE spectrum) a

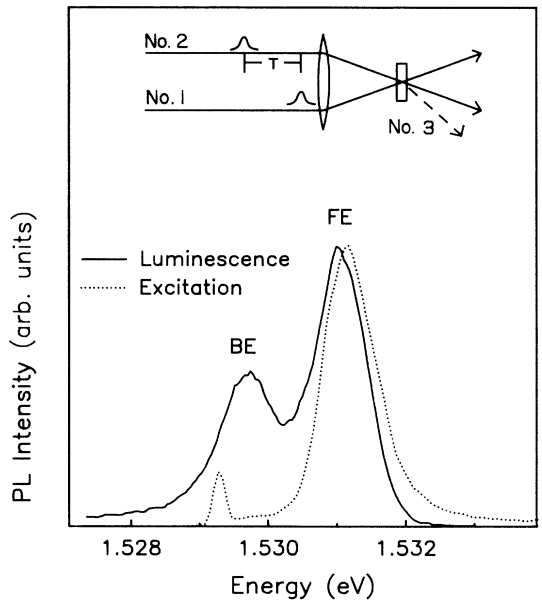

FIG. 1. Upper part: Schematical drawing of the experimental geometry. Lower part: Photoluminescence spectrum (solid line) and photoluminescence excitation spectrum (dotted line) in the vicinity of the lowest hh exciton transition, taken with a low-intensity cw laser at $T_L=2$ K. The small peak at 1.5292 eV in the PLE spectrum is scattered light from the laser.

second peak, whose energy is about 1.4 meV lower. This peak disappears at slightly elevated temperatures and saturates at high excitation densities. All these observations indicate a bound-exciton transition. The relative intensity of this peak compared to the hh free-exciton peak varies laterally across the sample.

The polarization decay of the excitons is traced by the two-pulse self-diffracted transient-grating technique. This technique was introduced to study the dephasing excitons in quantum wells by Schultheis, Sturge, and Hegarty[9] and is schematically depicted in the upper part of Fig. 1. The 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 T_2. The photons of the second beam are also diffracted by this transient grating into the direction $\mathbf{k}_3=2\mathbf{k}_2-\mathbf{k}_1$, where $\mathbf{k}_1$ and $\mathbf{k}_2$ are the wave vectors of the incoming beams. The decay of the polarization is reflected by the diffracted signal measured for varying delay time $T=t_2-t_1$ between the pulses. 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.[10]

Figure 2 shows the diffracted signal versus the time delay between the two pulses. The peak intensity of the excitation laser was chosen about 2.5 meV below the hh peak to avoid excitation of the lh transition. The upper three traces (solid lines) show the signal at low intensity $I_0\approx 160$ kW/cm^2 (corresponding to an exciton density of about 4×10^8 cm^{-2}) for lattice temperatures of $T_L=5$, 15, and 30 K. The decay of the diffracted signal for $T_L=5$ K clearly shows a modulation with a period of about 2.8 ps. This period corresponds to a splitting of 1.47 meV, in close agreement with the splitting between free- and bound-exciton transitions observed in the luminescence spectrum. The depth of modulation varies laterally across the sample, in agreement with the luminescence observations, which show a spatially varying bound-exciton luminescence intensity. Note that the rising wing ($T<0$) *does not show* a modulation, in contrast to the observation of light-hole and heavy-hole beats at negative delay time in another experiment.[6] This indicates that free and bound excitons show a weaker interaction compared to free excitons among themselves.[8] This observation agrees with bleaching experiments of free and bound excitons, where only the free excitons show a pronounced blueshift due to the many-body effects.

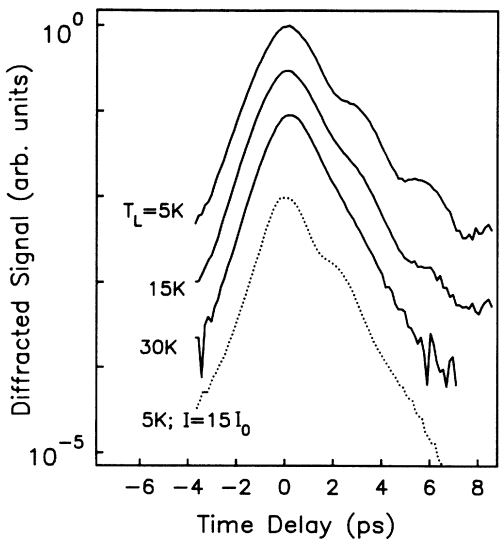

FIG. 2. Diffracted signal as a function of the delay time for low intensity ($I_0\approx 160$ kW/cm^2) and various lattice temperatures: $T_L=5$, 15, and 30 K (solid lines), and for $T_L=5$ K and higher intensity $15I_0$ (dotted line).

For higher lattice temperatures (traces for $T=15$ and 30 K in Fig. 2), the quantum beats disappear very quickly. However, if the phase relaxation time T_2 is similarly reduced[11] by increasing the exciton density ($I=15I_0$, dotted line in Fig. 2), the quantum beats are still visible. This difference could be caused by the rapid disappearance of bound-exciton transitions with increasing lattice temperature, in agreement with the luminescence measurements.

In conclusion, we report the observation of quantum beats between free- and bound-exciton transitions in GaAs/$Al_xGa_{1-x}As$ quantum wells. The beat frequency corresponds to the splitting between the excitonic transitions observed in luminescence spectroscopy.

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[1]See, for example, Y. R. Shen, *The Principles of Nonlinear Optics* (Wiley, New York, 1984).

[2]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).

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

[4]W. A. J. A. van der Poel, A. L. G. J. Sievers, and C. T. Foxon, Opt. Commun. **76**, 116 (1990).

[5]T. Tokizaki, A. Nakamura, Y. Ishida, T. Yajima, I. Akai, and T. Karasawa, *Technical Digest on Ultrafast Phenomena, 1990* (Optical Society of America, Washington, D.C., 1990), Vol. 6, p. 310.

[6]K. Leo, T. C. Damen, J. Shah, E. O. Göbel, and K. Köhler,

Appl. Phys. Lett. **57**, 19 (1990).
[7]B. F. Feuerbacher, J. Kuhl, R. Eccleston, and K. Ploog, Solid State Commun. **74**, 1279 (1990).
[8]K. Leo, M. Wegener, J. Shah, D. S. Chemla, E. O. Göbel, T. C. Damen, S. Schmitt-Rink, and W. Schäfer, Phys. Rev. Lett. **65**, 1340 (1990).
[9]L. Schultheis, M. D. Sturge, and J. Hegarty, Appl. Phys. Lett. **47**, 995 (1985).
[10]T. Yajima and Y. Taira, J. Phys. Soc. Jpn. **47**, 1620 (1979).
[11]L. Schultheis, A. Honold, J. Kuhl, K. Köhler, and C. W. Tu, Phys. Rev. B **34**, 9027 (1986).