Heavy-Light Hole Quantum Beats in the Band-to-Band Continuum of GaAs Observed in 20 Femtosecond Pump-Probe Experiments

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Coherent polarizations in the band-to-band continuum of bulk GaAs are studied in pump-probe experiments with 20 fs pulses. For the first time, we observe quantum beats on a 100 fs time scale that are due to an impulsively excited quantum coherence between heavy and light hole states. The beat frequency is determined by the heavy-light hole energy splitting changing continuously with the energy separation between the laser and the band gap. Theoretical calculations of the coherent response based on the semiconductor Bloch equations for a three-band scheme account for the data. [S0031-9007(96)02256-9]

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The fundamental nonequilibrium dynamics of optically excited semiconductors occurs on ultrafast time scales. Spectroscopy with femtosecond laser pulses provides direct information on such phenomena and has identified a coherent regime of material response in which the nonlinear polarization of the material and the electric field of the pulse couple in a phase-coherent way. In semiconductors with a direct band gap, both excitonic and free carrier excitations give rise to coherent polarizations with distinctly different properties [1–9]. Excitonic polarizations have been studied in great detail and new phenomena like wave packet propagation [1] and/or beating phenomena [2–4] have been observed in femtosecond four-wave-mixing experiments where the pulse spectrum overlaps with transitions of different frequency. For an ensemble with different transition frequencies, e.g., heavy hole excitons of different binding energy in quasi-two-dimensional (2D) structures, the oscillatory overall polarization originates from the contributions from the individual two-level constituents [2,4]. A different situation exists if the transitions are coupled via a common state and quantum interference in three-level systems causes polarization beats for heavy hole (HH) and light hole (LH) excitons [3–5]. In 2D systems, studies of the coherent response are facilitated by the—relatively slow—picosecond phase relaxation of excitonic polarizations.

Much less is known on nonlinear polarizations in the band-to-band continuum of semiconductors, showing dephasing kinetics in the sub-100-fs regime [6]. Recent four-wave-mixing experiments using sub-20-fs pulses close to the band gap of bulk GaAs [7] and bulk CdSSe [8] gave evidence of oscillatory coherent polarizations. In Ref. [7], the oscillations were attributed to the coherent coupling of interband transitions and LO (longitudinal optical) phonons, giving insight into non-Markovian quantum kinetics. The much higher oscillation frequency in CdSSe was ascribed to intervalence band quantum beats of free carriers. In such experiments, however, the pulse spectrum overlapped with the absorption edge of the material where (i) excitonic effects are important, and (ii) the strong variation of absorption across the pulse spectrum leads to nonlinear propagation effects [9] and/or detuning oscillations [10]. Thus, optical excitation well above the absorption edge is important to proof free carrier quantum beats. In this Letter, we report the first observation of heavy-light hole quantum beats in the band-to-band continuum of bulk GaAs. Temporally and spectrally resolved pump-probe experiments with 20-fs pulses give detailed insight into the properties of such impulsively excited intervalence band polarizations. The beat frequency is determined by the HH-LH energy splitting and changes—due to the valence band dispersion—continuously with the energy separation of the laser relative to the band gap. Damping of the quantum coherence is dominated by the inhomogeneous broadening within the manifold of the impulsively excited heavy-light hole transitions. The experimental results are reproduced by theoretical calculations based on the semiconductor Bloch equations which include a three-band scheme to calculate the nonlinear optical response.

In our experiments, we studied an undoped and an n-doped (doping concentration 9×10^{16} cm^{-3}) GaAs

sample clad between $Al_{0.4}Ga_{0.6}As$ layers. Both samples with a 0.5 μm thick GaAs layer were grown by molecular beam epitaxy. The stationary absorption spectrum of the undoped crystal is shown in Fig. 1(a) (solid line, lattice temperature $T_L = 8$ K) [11]. Pump-probe studies in the band-to-band continuum of the two samples were performed with bandwidth-limited 20-fs pulses at $E_{\text{pulse}} = 1.61$ eV (pulse center) from a mode-locked Ti:sapphire laser. The laser spectrum in Fig. 1(a) (dashed line) overlaps with a broad manifold of transitions from HH and LH to conduction band (CB) states but not with the excitonic absorption lines. After interaction with the sample the probe pulses are detected integrally or spectrally dispersed by a monochromator (spectral resolution 8 meV). Various energy separations between the band gap $E_{\text{gap}}(T_L)$ and the laser (E_{pulse}) were realized by varying the temperature T_L of the sample. The excited carrier density n_{ex} was estimated from the number of photons absorbed in the material per unit area. The data presented here were taken with $n_{\text{ex}} \simeq 3 \times 10^{15}$ cm^{-3} which is a factor of 30 below the electron concentration in the n-doped sample, and with parallel (linear) polarization of pump and probe.

In Fig. 1(b), we present time-resolved data for the undoped (solid line) and the n-doped sample (dashed line, $T_L = 8$ K). The spectrally integrated change of transmission is plotted versus the delay between pump and probe. The dash-dotted line gives the cross correlation of pump and probe pulses recorded at the sample location. The two transients show a steplike increase of transmission followed by a slow decay in the time window up to 180 fs. In Fig. 2(a) spectrally dispersed data at different positions E_{det} within the probe spectrum are shown [cf. arrows in Fig. 1(a)]. Close to the center of the pulse ($E_{\text{det}} = 1.60$ eV) the transients are similar to the spectrally integrated data. Above and below this value the data show—on top of the steplike bleaching—an oscillatory behavior at early times which is particularly strong for small E_{det}, e.g., at $E_{\text{det}} = 1.56$ eV. It is important to note that the dynamics in both GaAs samples, undoped and n-doped, is almost identical. Additional measurements

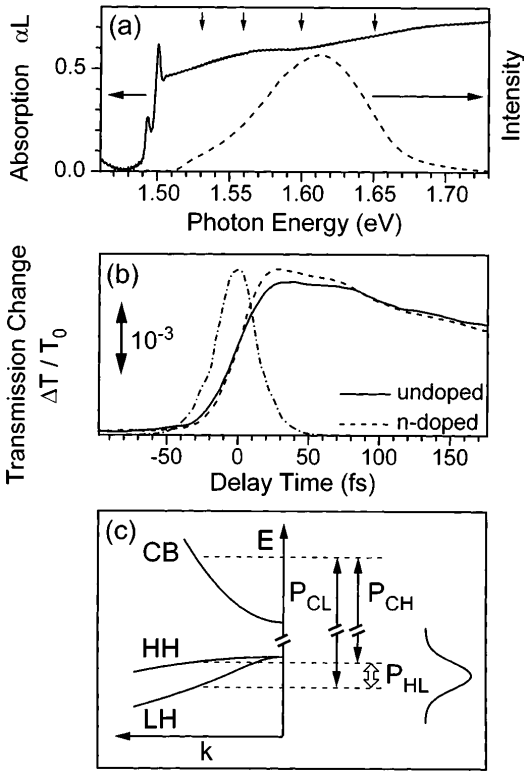

FIG. 1. (a) Steady state absorption αL of a $L = 0.5$-μm thick undoped GaAs sample at a lattice temperature of $T_L = 8$ K (solid line, α: absorption coefficient). Dashed line: spectrum of the 20-fs pulses. (b) Time-resolved transmission changes of the undoped (solid line) and n-doped (dashed line) GaAs sample for spectrally integrated probe pulses. The change of transmission $\Delta T/T_0 = (T - T_0/T_0)$ is plotted vs the delay between pump and probe (T, T_0: transmission with and without excitation). Dash-dotted line: cross correlation of pump and probe. (c) Schematic band structure of GaAs: conduction band (CB), heavy hole (HH), and light hole (LH) valence bands. Coherent polarizations (arrows) between various bands P_{CL}, P_{CH}, and P_{HL} occurring in the pump-probe experiments are shown for a single k state (three-level system). Impulsive excitation (Gaussian curve) of P_{HL} causes quantum beats.

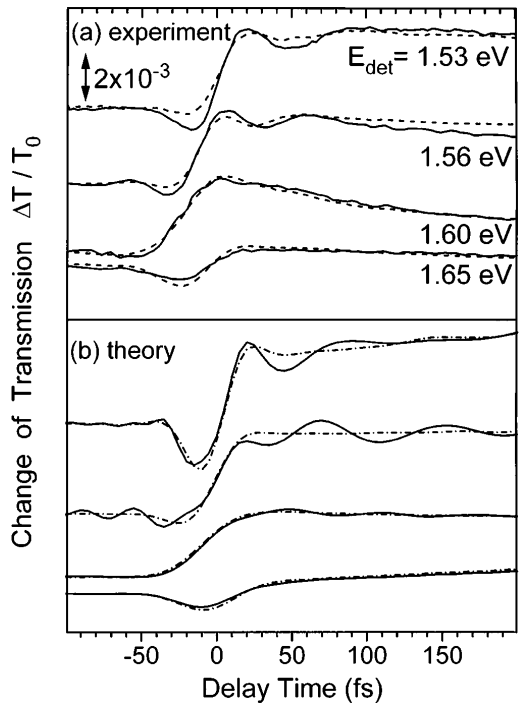

FIG. 2. (a) Time-resolved transmission changes of the undoped (solid lines) and n-doped (dashed lines) GaAs sample. The spectrally dispersed data (spectral resolution 8 meV) were taken at the spectral positions indicated by arrows in Fig. 1(a). Both samples give almost identical signals. (b) Simulation of the transient data in (a) based on the semiconductor Bloch equations for a three-band model. Solid and dashed lines: intervalence band polarization included and excluded, respectively.

showed that the oscillations disappear if the bandwidth of the laser pulses is reduced *before* interaction with the sample, and give insight into the polarization dependence of the oscillations. Oscillatory signals occur with parallel linear and cocircular polarization of pump and probe. They are absent with perpendicular linear and countercircular polarizations.

The oscillation period depends on the center frequency of the excitation pulses with respect to the band gap of GaAs. We tuned the excess energy $\Delta E_{\mathrm{ex}} = E_{\mathrm{pulse}} - E_{\mathrm{gap}}(T_L)$ by varying the lattice temperature T_L between 8 and 300 K. In Fig. 3, time-resolved data recorded at $E_{\mathrm{det}} = 1.56$ eV are shown for $\Delta E_{\mathrm{ex}} = 134$ meV [as Fig. 2(a)], 180, and 231 meV. The transients have a similar shape but the oscillation frequencies change with ΔE_{ex} as is evident from the dashed lines connecting points of constant phase. The oscillation frequencies $\nu_{\mathrm{osc}} = E_{\mathrm{osc}}/h$ (h: Planck's constant) were determined by a Fourier analysis of the data. In the inset of Fig. 3, the values of E_{osc} are plotted as a function of the excess energy ΔE_{ex} (squares), showing an almost linear dependence on ΔE_{ex}.

The ultrafast transmission changes observed in our spectrally and time-resolved experiments exhibit two contributions: (i) a steplike increase of transmission which rises within the time resolution of the experiment and shows a slow decay at later times, and (ii) a coherent oscillatory signal superimposed on component (i). The first signal is mainly due to state filling by electron-hole pairs and is strongly influenced by correlated many body effects, e.g., screening, among the photoexcited carriers. The generation process is influenced by the coherent coupling of the laser pulses and the material [12]. At later times, this bleaching represents an incoherent signal determined by the transient distribution of Coulomb correlated carriers. The redistribution of electrons and holes is governed by Coulomb and carrier-phonon scattering and will be discussed elsewhere. For an analysis of the coherent contribution, it is important to note that both pump and probe pulses overlap exclusively with the band-to-band continuum of GaAs [Fig. 1(a)]. Thus, transmission changes due to excitonic polarizations [1–3] or detuning oscillations between the exciton resonance and the laser field [10] do not occur. This conclusion is supported by the very similar results for the n-doped sample where excitonic polarizations are absent.

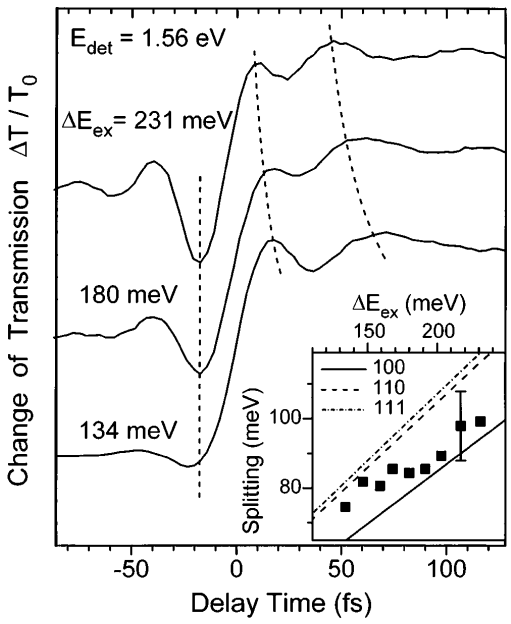

FIG. 3. Spectrally dispersed transmission changes of the n-doped sample at $E_{\mathrm{det}} = 1.56$ eV for different excess energies ΔE_{ex} of the pulse center at 1.61 eV relative to the band gap. The frequency of the quantum beats increases with ΔE_{ex} (the dashed lines connect points of identical phase). Inset: Oscillation energy of the quantum beats as a function of the excess energy ΔE_{ex}. Dashed and solid lines: calculated energy separation of the optically coupled heavy and light hole states vs ΔE_{ex} for different directions in k space.

The coherent signal found in our measurements is dominated by quantum beats in the band-to-band continuum. In Fig. 1(c), the underlying mechanism is explained schematically: The 20-fs excitation pulses couple to a manifold of transitions from HH and LH states to CB states in a large area of k space. Because of the very broad pulse spectrum, both transitions HH-CB and LH-CB for a single k vector are covered. This corresponds to a three-level situation with a common state in the conduction band, causing additional contributions to the third order nonlinear polarization $P^{(3)}$ which determines the differential transmission spectrum (pump probe minus probe only) in the limit of weak probe pulses [13]. The impulsive excitation of the coherent HH-LH polarization which oscillates with the difference frequency $(E_{\mathrm{HH}}\text{-}E_{\mathrm{LH}})/h$, leads to the oscillatory transmission changes which are particularly pronounced on the low-energy tail of the pulse spectrum [$E_{\mathrm{det}} = 1.56$ eV, Figs. 2(a) and 3].

Because of the lack of a quantization axis, the polarization dependence of HH-LH beats in bulk GaAs is more complex than for excitons in quantum wells. A theoretical analysis which is based on the polarization selection rules and will be discussed elsewhere predicts quantum beats of large amplitude for parallel linear and cocircular polarizations and much weaker beats with opposite phase for perpendicular linear and countercircular polarizations. This is in qualitative agreement with our data and, thus, supports our interpretation.

The change of beat frequency with the position of the excitation in k space gives strong independent evidence for the HH-LH quantum beats. The data in Fig. 3 are in agreement with the HH-LH splitting derived from an 8×8 $\mathbf{k} \cdot \mathbf{p}$ band structure calculation along the symmetric k directions {100}, {110}, and {111} (lines) [14]. The rapid damping of the oscillations on a 100-fs time scale is due to the following mechanisms: Transitions with

different HH-LH splittings couple to the spectrally broad pump and probe pulses and show a different time evolution, resulting in a loss of mutual phase coherence (inhomogeneous broadening). Furthermore, phase-breaking carrier-carrier and carrier-phonon scattering lead to the dephasing of the coherent HH-LH polarization. The moderate excitation density of 3×10^{15} cm^{-3} results in Coulomb scattering times among excited carriers of several hundreds of femtoseconds [12,15], i.e., long compared to the time interval during which quantum beats are observed. The very similar results for the undoped and n-doped sample show that interaction of excited carriers with the cold electron plasma in the n-doped sample makes a negligible contribution to the overall dephasing. Eventually, LO phonon scattering with time constants of the order of 150 fs limits the phase-coherent regime of optical response.

We performed theoretical calculations of the microscopic dynamics based on the semiconductor Bloch equations for an isotropic three-band model. The basic variables are given by the three distribution functions of electrons, heavy holes, and light holes, the two interband polarizations between the conduction band and the two valence bands, as well as the intervalence band polarization connecting HH and LH holes states with the same k vector which describes a coherent superposition of HH and LH states. The theory is formally equivalent to the multisubband case in quantum well structures as discussed in Ref. [16]. Coulomb correlation among the carriers was taken into account in the screened Hartree-Fock approximation by using a time-dependent static screening. Scattering processes due to carrier-carrier and carrier-LO-phonon interaction were treated on a microscopic level for all distribution functions, interband, and intervalence band polarizations. To account for the pump-probe geometry, a spatial Fourier decomposition was performed as has been used previously for the case of four-wave mixing experiments [17,18]. Details of the theoretical treatment will be published elsewhere. The calculated transmission changes are presented in Fig. 2(b). The amplitude and phase of the beats as well as the variation of beat frequency with the excess energy of the laser pulses are in very good agreement with the experiment. To verify the role of the HH-LH coherence, calculations have been performed where the intervalence band polarization has been switched off. As shown in Fig. 2(b) (dash-dotted lines), oscillations are absent in this case, a clear proof that they are caused by HH-LH quantum beats. Coulomb correlation results in an enhancement of both the coherent and incoherent transmission changes in the red part of the laser spectrum, in agreement with the results of our spectrally resolved measurements.

In summary, pump-probe experiments with 20-fs pulses gave evidence of impulsively excited heavy-light hole quantum beats in the band-to-band continuum of undoped and n-doped GaAs. The coherent oscillations persist up to delay times of 100 fs and show a frequency depending on the spectral position of the pulses with respect to the band gap. The observed beat frequencies are in good agreement with the respective energy splitting of the optically coupled heavy and light hole states. Theoretical calculations based on the semiconductor Bloch equations reproduce the experiments very well.

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