Coherent Dynamics of Excitonic Wave Packets

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We report on the coherent dynamics of excitonic wave packets observed in transient four-wave mixing experiments on strained InGaAs/GaAs quantum-well structures. The excitonic noneigenstates are created by using laser pulses shorter than the electron round trip time within the exciton orbital. A characteristic temporal modulation of the diffracted signal is observed, which resembles the coherent dynamics of an electron wave packet in a hydrogen atom expected for the case of a simultaneous excitation of all bound and the lowest continuum states.

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Recently, localized electronic wave packets have been created in the electronic shell of atoms by the simultaneous excitation of several atomic Rydberg states using short optical pulses [1–3]. The coherent dynamics of these electronic noneigenstates have been detected by performing field-ionization experiments [1] as well as optical pump and probe experiments [2].

In semiconductor physics, the concept of excitons, i.e., of electron-hole pairs held together by the attractive Coulomb interaction, corresponds to the problem of a hydrogen atom in atomic physics. In particular, quantum-well structures, where excitonic effects are enhanced, clearly reveal exciton spectra with a hydrogenic series [4, 5]. Analogously to the creation of wave packets in atomic shells, the bound and unbound states of an electron-hole (e-h) pair associated with a single one-particle interband transition could also be considered as a basis for the creation of an excitonic noneigenstate. In a simple-minded picture, a spectrally broad laser pulse should allow the coherent superposition of various bound states $(1s, 2s, \ldots)$ and unbound e-h scattering states (see excitation scheme in Fig. 1). In other words, if the excitonic transitions were excited by a laser pulse which is shorter than the classically expected orbit time τ_{orb} of the electron around the hole, a coherent electron-hole wave packet is expected to be created developing in time according to the time-dependent Schrödinger equation.

Yet, the exciton problem in semiconductors is different compared to the case of hydrogen atoms in two important aspects: First, the concept of an exciton is a many-body problem. The corresponding many-body Hamiltonian can be transformed into an effective two-particle hydrogenlike Hamiltonian by making several assumptions, e.g., by neglecting exchange terms [6]. It has recently been shown that the inclusion of many-body terms beyond the Wannier equation is important for a complete understanding of coherent optical experiments on excitons in semiconductors [7–11]. Second, in atoms, the selection rules only allow optical transitions between atomic states, where the angular momentum of the electron is changed by ± 1. In semiconductors, this rule also applies for the Bloch part $[u_k(r)]$ of the electron wave function. An additional "exciton" selection rule states that only s-like exciton states can be optically excited [6]. Accordingly, optically generated excitonic wave packets are composed of hydrogenlike eigenstates of a different nature as compared to atomic wave packets.

During the last few years, the study of optically cre-

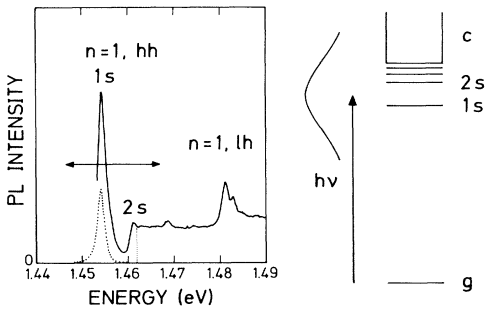

FIG. 1. Photoluminescence spectrum (dotted line) together with a photoluminescence excitation spectrum (solid line) of the MQW sample at $T = 5$ K. The dotted vertical line indicates the onset of the continuum transitions. The horizontal arrow shows the spectral width (FWHM) of the laser pulses. A schematic representation of the laser excitation process in the multilevel system is shown on the right-hand side (g =ground state, c =continuum states).

ated electronic wave packets in semiconductors has become possible by performing quantum beat experiments with short laser pulses [12–16]. Yet, in all of these studies the initially excited noneigenstate is formed by superimposing $1s$ exciton states related to *different* one-particle transitions. So far, the coherent dynamics of excitonic noneigenstates originating from the *same* single-particle transition has been an open question in the field of coherent laser spectroscopy in solids.

In this Letter, we report on transient four-wave mixing (FWM) experiments on high-quality $In_xGa_{1-x}As/GaAs$ quantum-well structures using subpicosecond laser pulses. Only the bound and unbound states of the lowest ($n = 1$) electron–heavy-hole Coulomb pair are excited. The diffracted signal exhibits characteristic modulations, which arise from the temporal development of the coherently excited electron-hole wave packet. Theoretical considerations show that, at low density, many-particle Coulomb interactions do not alter the coherent dynamics of the excitonic wave packet substantially. Under these conditions, the coherent dynamics of exciton noneigenstates closely resemble those of electronic wave packets in atoms, yet, with the difference that *all* bound as well as the lowest unbound states are excited simultaneously. This situation cannot be achieved in the case of atoms because the atomic binding energy is too large.

We have investigated $In_xGa_{1-x}As/GaAs$ single quantum-well structures as well as a multiple quantum-well structure. All samples were grown by molecular-beam epitaxy on undoped (100) oriented GaAs substrates [17]. Sample A contains 20 $In_{0.08}Ga_{0.92}As$ quantum wells of 10 nm width separated by 40 nm wide GaAs layers to avoid coupling between adjacent quantum-well states. Sample B contains single quantum wells of 30 nm and 10 nm $In_{0.12}Ga_{0.88}As$ separated by 200 nm thick GaAs barrier material [5]. The results of the FWM experiments are the same for all of these samples [18]. In the following, we therefore present only the experimental findings for the multiple quantum-well (MQW) sample A.

Figure 1 depicts the low-temperature photoluminescence (PL) spectrum as well as the photoluminescence excitation (PLE) spectrum of the MQW sample. The detection energy for the PLE measurement has been chosen to lie at the low-energy part of the $n = 1$ heavy-hole exciton line. The linewidth of the lowest $n = 1$ heavy-hole exciton transition amounts to 1.7 meV FWHM. The Stokes shift between the heavy-hole exciton emission line (PL) and the corresponding line in absorption (PLE) is less than 0.2 meV. In the PLE spectrum, the lowest $n = 1$ light-hole exciton transition is found at 1.4813 eV, i.e., at much higher energy than the $n = 1$ heavy-hole exciton. This large blueshift of the light-hole exciton is mainly due to the inherent strain within the InGaAs layers. Accordingly, it is possible to coherently excite only the heavy-hole exciton states without the simultaneous excitation of the light-hole exciton states.

The PLE spectrum not only shows the $n = 1$ heavy-hole 1s exciton at 1.4544 eV but also the $2s$ exciton transition at 1.4611 eV. This $2s$ excitonic transition is separated by 6.7 meV from the $1s$ exciton peak but it merges with the onset of the free-electron–heavy-hole continuum states. Assuming two-dimensional excitons, we can directly determine the exciton binding energy from the $1s$-$2s$ splitting [19]. We obtain a binding energy of 7.5 meV, which implies that the continuum states start at 1.4619 eV as indicated by the dotted line in Fig. 1. The classically expected orbit time τ_{orb} of an electron within the $1s$ heavy-hole exciton having a binding energy of 7.5 meV can be estimated to be about 300 fs. This means that laser pulses shorter than 300 fs or spectrally broader than 7.5 meV are required in order to create electron–heavy-hole wave packets in the InGaAs/GaAs MQW sample.

We use a mode-locked Ti-sapphire laser emitting linearly polarized light pulses of 110 fs duration. The spectral width corresponds to 20 meV (FWHM) as illustrated by the horizontal arrow in Fig. 1. The transient FWM experiments are performed in a two-pulse self-diffraction geometry [20]. The sample is excited by two subsequent laser pulses with parallel polarization and wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$ at times $t = 0$ and $t = \tau$, respectively. We then detect the FWM signal, diffracted into the direction $2\mathbf{k}_2 - \mathbf{k}_1$, time integrated as a function of the time delay τ between the pulses, using a slow photodetector. In the following, FWM experiments using time-integrated detection are called "transient," whereas FWM experiments with time resolution of the diffracted signal are called "time resolved." All experiments are performed at $T = 5$ K.

In Figs. 2(a)–2(c), the normalized diffracted FWM signal is shown as a function of the time delay τ and for three different spectral positions of the excitation laser pulses. The central photon energy is tuned 5 meV below (a), in resonance with (b), and 10 meV above the $1s$ heavy-hole exciton transition (c). The generated carrier

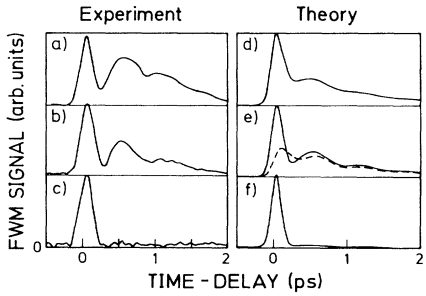

FIG. 2. FWM signal as a function of the time delay between the two laser pulses. The experimentally determined traces are shown on the left side for central photon energies detuned by $\Delta = -5$ meV (a), 0 meV (b), and +10 meV (c) from the $1s$ exciton transition. The calculated FWM curves using a simple multilevel system are shown as solid lines on the right side again for detunings of $\Delta = -5$ meV (d), 0 meV (e), and +10 meV (f). The dashed curve in (e) is the result of a calculation neglecting all continuum states.

density amounts to 4×10^9 cm^{-2}. The common feature of all three curves is the initial fast response of the signal with a maximum at $\tau = 70$ fs. For low-energetic as well as resonant excitation [Figs. 2(a) and 2(b)], the signal recovers and reaches a second maximum at $\tau = 550$ fs. In Fig. 2(a), a weak periodic modulation with a time period of about $T_B = 600$ fs is seen on the decaying FWM signal which corresponds to an energetic separation of $\Delta E = h/T_B = 6.9$ meV, which is in good agreement with the spectrally determined $1s$-$2s$ splitting.

In the following, we show that this characteristic temporal and spectral behavior of the transient FWM signal is a direct consequence of the simultaneous excitation of the bound exciton states $(1s, 2s, \ldots)$ and the unbound electron-hole continuum transitions. Realistic calculations should rely on numerical solutions of the full semiconductor Bloch equations [21]. However, we first start with a simplified model in order to understand more easily the origin of the observed unique quantum beat behavior. In a two-particle picture, the optically coupled bound and unbound exciton states form a multilevel system sharing a common ground state [6] as schematically shown in Fig. 1. Accordingly, we model the bound and unbound exciton transitions by assuming one ground state and N excited states. We then use the formalism of Ref. [22] to calculate the transient FWM signal for laser pulses of 110 fs duration. Our calculations include effects from finite pulse overlap. The energetic positions of the $1s$ and $2s$ excitons and of the continuum transitions as well as their respective optical dipole matrix elements are chosen to fit the measured absorption spectrum shown in Fig. 1. The continuum is modeled by closely spaced discrete levels. The results do not depend on this energy spacing if it is chosen less than about 1 meV. For the $1s$ and $2s$ bound exciton transitions the dephasing times are chosen to be $T_2 = 2$ ps and $T_2 = 1$ ps, respectively. These dephasing times were determined in an independent transient FWM experiment using spectrally narrow picosecond laser pulses [23]. We assume a constant dephasing time of $T_2 = 400$ fs for all unbound electron-hole continuum states [24]. The temporal evolution of the FWM signal is computed for the same spectral positions as chosen for the experimentally determined FWM curves shown in Figs. 2(a)–2(c). The results are shown as solid lines in Figs. 2(d)–2(f). It is seen that all qualitative features of the experimentally determined curves are reproduced. In Figs. 2(d) and 2(e), weak periodic modulations are observed on the slowly decaying FWM signal, which are due to quantum beats between the optically strong $1s$ and the optically weak $2s$ exciton transitions. In order to find the physical origin of the fast initial decay and the subsequent pronounced minimum of the FWM signal we have calculated the FWM curve for the same parameters as used for the solid line in Fig. 2(e) but without taking into account optical transitions into continuum states. The result is shown as the dashed line in Fig. 2(e). We obtain a beating between the $1s$ and $2s$ excitonic transitions, but the fast initial decay is absent, and the first minimum is less pronounced. This definitely proves that the fast initial FWM signal is due to excitation of continuum states. The shape of the FWM signal was also calculated taking into account only the $1s$ exciton and the continuum states. The result (not shown here) shows that the pronounced minimum, which is seen in Fig. 2(e) after the initial fast response, is due to destructive interference between the $1s$ exciton and the resonantly excited continuum states [25]. This minimum is even more pronounced in the experimentally determined FWM curves of Figs. 2(a) and 2(b), which is probably due to the fact that the dephasing times of the electron-hole continuum states are energy dependent and are perhaps longer than 400 fs for the conditions of our experiment. Our calculations show that a longer dephasing time of the continuum states enhances the destructive interference effect and thus diminishes the FWM signal in the first minimum. Altogether, this simple analysis shows that the characteristic modulations observed in the temporal evolution of the FWM signal reflect the coherent dynamics of an excitonic noneigenstate, i.e., of a Coulomb correlated electron-hole wave packet.

Recently, it has been shown that coherent exciton-exciton interactions might influence the time behavior of transient FWM signals; for high intensity, the transient FWM signal may exhibit two temporal maxima [7]. We also observe this behavior when the excitation density is increased by more than 2 orders of magnitude as compared to the density used in the experiments shown in Fig. 2. For low densities, it has been shown [8–11] that many-body Coulomb effects influence the real-time behavior of FWM signals measured in a time-resolved FWM experiment. In order to decide whether such effects also influence the *beating* behavior of the time-integrated (transient) FWM signal originating from the dynamics of excitonic wave packets, we have performed numerical solutions of the full semiconductor Bloch equations [21] for the case of a single quantum well, considering the present FWM geometry. The low-intensity absorption spectrum shown in the insets of Fig. 3 is obtained for Lorentzian homogeneous broadening corresponding to a dephasing time of 1 ps in the semiconductor Bloch equations. Transient FWM signals are shown in Figs. 3(a) and 3(b) for detuned ($\Delta = -4.2$ meV) and resonant ($\Delta = 0$ meV) excitation, respectively, using equal Rabi frequencies of 0.42 meV for both pulses. Again, destructive interference between $1s$ and continuum states causes a fast initial decay of the FWM signal which evolves into periodic modulations for larger time delays. The beats become more pronounced with decreasing detuning and *reveal essentially the same features as obtained from the noninteracting multilevel model.* A direct consequence of many-body Coulomb effects [7] is an additional modulation of the transient FWM signal at negative time delays. As already mentioned above, we observe such a behavior for higher excitation intensities.

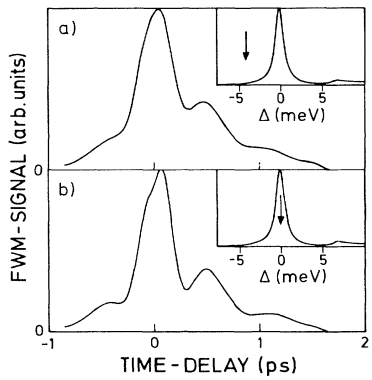

FIG. 3. Calculated FWM curves as a function of time delay obtained by numerically solving the semiconductor Bloch equations. As indicated by the arrows in the corresponding absorption spectrum (shown as insets), the central photon energies are detuned by $\Delta = -4.2$ meV (a) and 0 meV (b) from the $1s$ exciton transition.

Altogether, many-body effects definitely alter rise and decay times of FWM signals [10, 11], but they do not substantially alter the temporal positions of constructive and destructive interference in quantum beat experiments. Thus, it is the basic dynamics of electron-hole wave packets which is observed in a transient FWM experiment.

Finally, our results indicate that, in general, an initially fast decaying FWM signal may not always be due to fast dissipative dephasing processes, but can also be caused by destructive interference phenomena. This is particularly true for subpicosecond FWM experiments when partial excitation of continuum states cannot be avoided.

In summary, we have performed subpicosecond FWM experiments on InGaAs/GaAs quantum-well structures. We observe characteristic temporal modulations of the FWM signal. We have shown that they reflect quantum beating between bound and unbound electron–heavy-hole transitions by analyzing a simple multilevel system. Accordingly, the transient FWM experiment directly monitors the coherent development of the optically excited electron-hole wave packet. In addition, we have demonstrated theoretically that at low carrier densities many-body effects do not alter the temporal development of the excitonic noneigenstate substantially. Consequently, the coherent exciton dynamics in these semiconductor quantum wells closely resemble the dynamics of electronic wave packets in atoms.

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