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ULTRAFAST MAGNETOEXCITON DYNAMICS IN GaAs

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Ultrafast Magnetoexciton Dynamics in GaAs

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Abstract:

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Using ultrashort pulsed wave mixing spectroscopy, we investigate coherent emission from magnetoexcitons in GaAs. By spectrally resolving the emission, we relate the non periodic beat observed temporally to quantum interferences, which we attribute to Coulomb interactions between magnetoexcitons. Contrary to experiments in quantum wells, these interactions persist for field strengths up to 10T.

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Ultrafast Magnetoexciton Dynamics in GaAs

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A bulk semiconductor provides an excellent system for optically studying the ultrafast dynamics of Coulomb correlated magnetoexcitons. The application of a magnetic field to a 3-D system quantizes the energy levels of states with momentum perpendicular to B and. in contradistinction to experiments performed on 2-D quantum well systems, leaves a 1-D continuum of states with momentum parallel to B. The presence of the continuum states has a dramatic influence on both the linear and nonlinear optical properties. Thus, the.experiments described below track the evolution of the semiconductor from a 3-D to a 1-D system and reveal surprising optical properties not observed in the previously reported studies in quantum wells.

In GaAs, under the application of a magnetic field, the discrete higher order magnetoexciton states are energetically degenerate with the 1-D. continuum of states of the lowest order magnetoexciton. The configuration interaction between an isolated resonance superimposed energetically upon a continuum of states was described theoretically by Fano. [I] The resulting state is a superposition of the original discrete state and the continuum of states and the resulting resonance has a distinctive asymmetric lineshape due to quantum interference. Asymmetric Fano resonances have been observed in the linear absorption spectrum of GaAs under magnetic fields. [2] Our present experiments investigate both the absorption properties of these states and also the ultrafast coherent emission from these resonances.

Figure 1: GaAs linear absorption at 1.6K for several magnetic field strengths.

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Fig. 1 shows the linear absorption spectrum of our GaAs sample at 1.6K taken with circularly polarized light for several magnetic field strengths. At B=0T, we see that due to strain the light hole (LH) exciton and the heavy hole (HH) exciton are split, with the LH exciton at lower energy. [3] By increasing the field strength to 12T, we verified that the resonances observed in the spectrum form a fan chart pattern and do indeed correspond to two families of magnetoexcitons. The most interesting feature here is the asymmetric lineshape of the higher order magnetoexciton resonances. Immediately at 2T the resonances in the continuum show these features. As the field strength increases to 6T, say, the resonances become more pronounced. We attribute this change in profile to a field dependent coupling between the discrete state and the underlying continuum of states. Finally, at the highest field strengths, the magnetoexciton states are separated well enough to excite an isolated resonance with our 1 OOfs laser pulse. In the nonlinear optical experiments described below, we concentrate on the ultrafast dephasing properties of the lowest lying magnetoexciton states.

Four wave mixing (FWM) experiments were performed using 100fs pulses from a Ti:Sapphire laser tuned to excite the lowest order HH and LH magnetoexcitons. The beam was split into two co-circularly polarized beams, propagating in directions k_1 and k_2 , with pulses variably time delayed by Δt . The beams overlapped spatially on our sample, which was held at 1.6K inside of a split coil magnet-cryostat. The FWM signal emitted in direction $2k_1-k_2$ was then either time-integrated (TI), i.e. its intensity was measured vs. At, or for a fixed At the spectrum of the FWM emission was measured.

Figure 2: TI-FWM for B=10T (top), 6T, 2T, and 0T (bottom).

The TI FWM signal is shown in Fig. 2 for various field strengths and at excitation density $N_x \approx 10^{16}$ cm⁻³. At zero field, there is a nonexponential decay in approximately Ips for positive time delays and otherwise the lineshape is featureless. With field, however, the temporallineshape is completely different. There is an obvious beating modulation on the TI signal which changes significantly with increasing field strength. Importantly, this modulation is excitation density dependent and becomes much less pronounced at higher densities. Moreover, this non periodic beating is observed for both positive and negative At. These are indications of strong Coulomb interactions between magnetoexcitons. These interactions have been studied extensively for 2-D systems, where theoretical models [4] demonstrate that they are at the origin of these distinctive TI lineshapes. Fundamentally different from 2-D systems, where the interactions become

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quenched at high fields, our measurements indicate that in 3-D these features persist for fields as high as 10T. This result is understood in terms of the 3-D linear data, which demonstrate a strong coupling between states for fields as high as 12T.

In order to better understand the TI beat modulation, we measured the spectrum of the FWM emission for each of several Δt for a 6T magnetic field and $N_x \approx 10^{16}$ cm⁻³. Fig. 3 shows the measured TI profile and the evolution in time of the FWM spectra around a beat node. It is clear that the competition between spectral components results in the beating observed temporally. From these data we also measure the dynamics of each component individually. We find that as these components evolve in time, they decay and/or beat asynchronously with distinct dephasing times. For example, the component at the HH exciton $(\lambda=818$ nm) decays slowly (in at least 1.1ps) and exhibits a beating modulation, whereas a higher energy component decays very rapidly, in about 90fs. Thus, it is the complex interplay spectrally which manifests itself as non periodic beats in the time domain.

Figure 3: For B=6T, TI-FWM (top) and FWM Power Spectra (bottom) for $\Delta t = -560$ *fs, -400fs, -320fs, .. 240ft, and -160/s.*

Experiments are in progress to investigate the microscopic origin of these various spectral components. From the magnetic field and excitation density dependent TI-FWM measurements, it is clear that Coulomb interactions between magnetoexcitons strongly affect the emission properties of the system. We attribute the observed spectral properties to these interactions and hope these results will stimulate the development of a model for ultrafast magnetoexciton dynamics in 3-D systems.

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