Observation of the ultrafast two-photon coherent biexciton oscillation in a GaAs/$Al_xGa_{1-x}As$ multiple quantum well

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(Received 13 May 1996)

Polarization-dependent heterodyne four-wave-mixing measurements with a phase-conjugate Michelson interferometer reveal a macroscopic nonradiative 770-THz oscillation in the nonlinear optical susceptibility of GaAs multiple quantum wells. The oscillation is interpreted as the temporal evolution of the two-photon nonradiative biexcitonic coherence. [S0163-1829(96)50932-X]

The role of the biexciton in semiconductor optical interactions has been studied intensely for over two decades, particularly in wide-gap materials. Extension of this work to GaAs-based materials has been a relatively recent development, complicated by the relatively small biexciton binding energy. Observation has been difficult even in multiple-quantum-well (MQW) structures, for which the binding energy is typically enhanced by an order of magnitude over the bulk value. The first report of a biexciton in a GaAs/$Al_xGa_{1-x}As$ MQW was made by Miller,[1] who obtained a heavy-hole biexciton binding energy of approximately 1 meV in luminescence. This value was soon corroborated by variational calculations.[2] Since then, luminescence has confirmed many properties of the biexciton in GaAs MQW's.[3–8] Most recently, detailed thermodynamic[9] and polarization[10] studies have underscored the importance of biexcitonic processes. Although detailed agreement with theory cannot be claimed generally, many basic properties of biexcitons in GaAs are now well established.

The first indication of a biexcitonic nonlinearity in GaAs MQW's was discovered recently by Feuerbacher *et al.*[11] in self-diffracted four-wave mixing (FWM). Vigorous discussion of the details of biexcitonic nonlinearities in GaAs has ensued.[12–21] The biexcitonic nonlinear response in FWM arises from two competing and fundamentally different excitations paths that have analogs in the theory of three-level atoms.[22] The path that dominates in studies of biexcitons in long-time-scale measurements (e.g., luminescence) is an incoherent stepwise (SW) generation of biexcitons excited directly from a population of excitons. The other path excites biexcitons through a totally coherent excitation[23] via a nonradiative two-photon coherence (TPC) induced between the ground and biexciton state, which oscillates at the biexciton frequency $\Omega_{bg}=2\Omega_{eg}-\Delta$, where $\hbar\Delta$ is the biexciton binding energy. This path becomes important for time scales comparable to the TPC dephasing time. Here, we qualitatively demonstrate the unique features of the TPC path in GaAs by using heterodyne FWM to identify the ultrafast 770-THz TPC oscillation. The utility of interferometric measurements in the vicinity of a single-photon resonance has been established in recent measurements of nonlinear phase shifts[24,25] and pulse distortions[26,27] in GaAs MQW's. Identification of the TPC, however, requires a very different interferometer specifically designed for investigating two-photon resonances, as described below and shown in Fig. 1.

Detecting the TPC requires careful discrimination of several signals, especially contributions from two-level FWM, including concomitant exciton-exciton interactions (EEI), which partially relax nominal time ordering. In phase-conjugate FWM, two pulses, $E_1(\omega,\mathbf{k}_1)$ and $E_2(\omega,\mathbf{k}_2)$, interact with the sample at times t_1 and t_2, respectively, to produce an excitation grating. A third pulse, $E_3(\omega,-\mathbf{k}_2)$, diffracts from the grating to produce a signal polarization propagating in the background-free direction $-\hat{\mathbf{k}}_1$. Except as noted, we take the arrivals of E_2 and E_3 to be simultaneous, $t_3=t_2$, a time $\tau\equiv t_2-t_1$ after E_1. The appearance of a signal for negative delays, $\tau<0$, is evidence of EEI such as local fields,[28] excitation-induced dephasing,[29] and (in three-level systems) the TPC, which has no grating analogy. In the data below, we demonstrate the ultrafast TPC oscillation and present polarization-dependent data to assess the influence of non-TPC EEI on the experiment.

A density matrix representation illuminates key features of the SW and TPC paths as manifested in FWM; subscripts denote *g*round, *e*xciton, and *b*iexciton amplitudes. The noninteracting two-level FWM signals ($\tau>0$) are represented in the rotating wave approximation by the perturbation sequence

$$\rho_{gg}\xrightarrow{E_1^*}\rho_{ge}\xrightarrow{E_{2,3}}\rho_{ee}\xrightarrow{E_{3,2}}\rho_{eg}.$$

(The notation emphasizes that the time ordering of E_2 and E_3 is irrelevant, even if E_2 and E_3 are not coincident.) The SW biexcitonic pathway ($\tau>0$) is

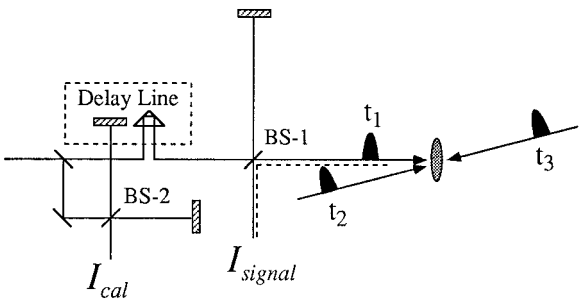

FIG. 1. Geometry of the phase-conjugate Michelson interferometer.

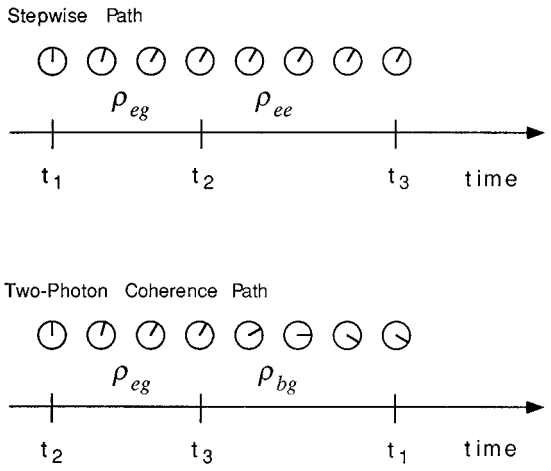

FIG. 2. Schematic time evolution of selected density matrix elements for the SW and TPC biexcitonic FWM signals.

$$\rho_{gg} \xrightarrow{E_1^*} \rho_{ge} \xrightarrow{E_{2,3}} \rho_{ee} \xrightarrow{E_{3,2}} \rho_{be},$$

and the TPC pathway ($\tau<0$) is

$$\rho_{gg} \xrightarrow{E_{2,3}} \rho_{eg} \xrightarrow{E_{3,2}} \rho_{bg} \xrightarrow{E_1^*} \{\rho_{be}, \rho_{eg}\}.$$

A close inspection of the two-level, SW, and TPC paths reveals a crucial distinction. The population, ρ_{ee}, which contributes to the two-level and SW paths, is quasistationary, but the electronic phase of the TPC evolves via the ultrafast quantum coherence, $\rho_{bg} \sim \exp(-i\Omega_{bg}t)$, after the arrival of the first two pulses. The TPC is independent of any exciton population. This is illustrated in Fig. 2, where a nonessential delay, $t_3 - t_2$, has been introduced only for exposition. Relaxation of the TPC represents a loss of coherence between the ground and biexcitons states and is distinct from exciton dephasing, which reflects the homogeneous exciton linewidth. Although the TPC is nonradiative (i.e., direct biexciton creation is not dipole allowed), the phase of the TPC is sampled by E_1 and impressed upon the radiative signal polarizations, ρ_{be} and ρ_{eg}. The TPC can therefore be detected interferometrically by heterodyne mixing of the FWM signal with a local oscillator, E_r. The total signal measured by a slow detector is modulated at the TPC frequency with respect to delay τ. This principle was used by Kuwata-Gonokami, *et al.*, to study CuCl.[30]

We employ the femtosecond phase-conjugate Michelson interferometer in Fig. 1 to observe the TPC as a function of the delay, τ. Since this method detects a phase shift, not the envelope of the emission, pulses need not be short on this time scale, and we use picosecond pulses to avoid hh-lh beats (where hh and lh are heavy hole and light hole, respectively). A single synchronously pumped mode-locked dye laser tuned to 800 nm provides approximately Gaussian pulses with 4 ps autocorrelation width and 0.7 meV bandwidth resonant with the hh1 exciton resonance at 5 K. The MBE-grown sample contains ten 100-Å GaAs wells separated by 100-Å $Al_{0.3}Ga_{0.7}As$ barriers. The absorption line width is 1.0 meV, and the Stokes shift is 0.3 meV. The main interferometer (using BS-1 in Fig. 1) is a modified Michelson design;

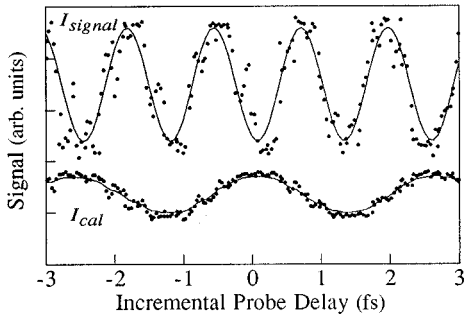

FIG. 3. Typical simultaneous scans (offset for clarity) of signal and calibration interferometers for $\tau \approx -3$ ps. Lines are guide to the eye. The approximate 2:1 ratio of the TPC and laser (resonant with the exciton) is clear.

the sample acts as a phase-conjugate mirror forming one arm of the interferometer. The delay line is dithered over several optical cycles using a piezoelectric element. This special geometry, in which E_r and E_1 are delayed in equal increments, assures that the interferogram is sensitive only to the delay, $\tau = t_2 - t_1$, and not to the time interval, $t_1 - t_r$. In contrast to earlier interferometric studies, which were not sensitive to the temporal oscillation of the TPC, this geometry produces no interference fringes near the laser frequency, independent of the origin of the nonlinear response. An auxiliary Michelson interferometer (using BS-2) produces conventional laser fringes to simultaneously calibrate the delay line.

A typical single scan is displayed in Fig. 3, with solid lines to guide the eye. The scatter is due to small laser frequency instabilities affecting the FWM signal. The (approximately) two-to-one ratio between the signal and calibration periods is apparent, and ensemble averaging gives a beat period of approximately 1.3 fs, corresponding to a 770-THz TPC oscillation. The 10% uncertainty precludes a measure of the biexciton binding energy, which would require precision of 0.03%. As indicated above, interpretation of this data in terms of the TPC oscillation requires care to eliminate potential alternative origins of the oscillation. For example, the two-level subsystem comprising only the exciton and ground states produces a nearby competing oscillation at $\omega + \Omega_{eg}$, via non-TPC EEI as well as finite pulse width effects. These signals arise from ''ordinary'' interferometry of two-level FWM, rather than from the electronic phase of the TPC.

Signals induced by EEI generally introduce some ambiguity into *all* time orderings. For example, a class of negative-delay EEI signals described by

$$\rho_{gg} \xrightarrow{E_{2,3}} \rho_{eg} \xrightarrow{E_1^*} \rho_{ee} \xrightarrow{\overline{E}_{3,2}} \rho_{eg},$$

where $\overline{E}_{2,3}$denotes a field (E_2 or E_3) which actually arrives *first* but persists in the sample via EEI, might be confused with the TPC. These ''local field'' signals are strongly suppressed by polarization interference in our strongly inhomogeneously broadened sample.[29] Local fields decay with the inverse *inhomogeneous* linewidth, T_2^*. The TPC signal, however, decays with the inverse *homogeneous* linewidth, T_2. For our MQW, $T_2^* \ll T_2$.

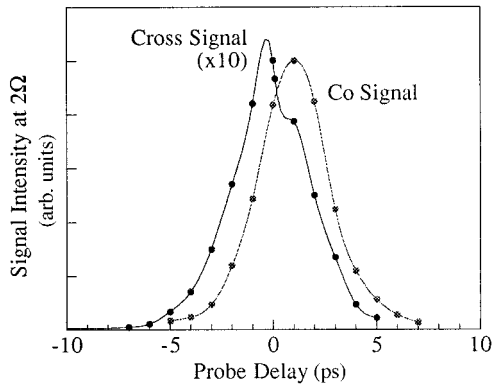

FIG. 4. Intensity of the approximately $2\Omega_{eg}$ oscillation for co- and cross-polarized fields versus delay, $\tau=t_2-t_1$. The cross-polarized signal favors negative delay, as expected for the TPC. Lines are guides to the eye.

Finite pulse width effects, however, may be significant if the pulse width is comparable to the FWM decay time, as in our experiment. But even for moderately pulse-width-limited data, the non-TPC signals favor positive rather than negative delay. Moreover, the signal for $\tau>0$ has a strong polarization dependence in GaAs MQW's and is substantially reduced for E_1 cross-linearly polarized with respect to both E_2 and E_3, $E_1 \| E_r \perp E_{2,3}$. In Fig. 4 we plot the Fourier spectral power density of the heterodyne signal in the vicinity of Ω_{bg} for both co- and cross-linearly polarized configurations, as a function of delay, τ. For cross-polarized fields, the oscillation favors negative delay, as expected for the TPC. This observation strongly supports the interpretation that the negative-delay signal is in fact the TPC.

For completeness, we note that the spectrally resolved FWM reveals two peaks: a ''free'' exciton peak and a Stokes-shifted peak, similar to an earlier observation.[20] In our experiment, however, the relationship between the 0.5-meV Stokes shift and the biexciton binding energy is complex, since the ''free'' exciton emission is dominated by excitons localized by disorder. More generally, experimental determination of the biexciton binding energy is complicated by strong inhomogeneous broadening. Therefore, spectrally resolved FWM does not provide a simple determination of the biexciton binding energy in this sample.

The observation of the TPC oscillation shows the presence of the nonradiative biexciton coherence and demonstrates its importance for observation time scales comparable to the TPC dephasing time. Continued refinement of techniques to quantitatively characterize the TPC oscillation will enable measurements of coherent nonradiative biexciton dynamics not accessible in conventional optical measurements such a luminescence, where the relative contribution of the TPC is generally negligible compared to the incoherent SW contribution.

The authors thank Professor M. Kuwata-Gonokami for helpful discussions and for sharing his work on CuCl prior to publication. This work was supported by the AFOSR, ARO, and NSF.

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