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Multiple-quantum 2D spectroscopy of many-body correlations in GaAs quantum wells

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ABSTRACT
Multiple-quantum two-dimensional Fourier transform optical (2D FTOPT) spectroscopy was developed and conducted on GaAs quantum wells. Spatiotemporal femtosecond pulse shaping was used to control the optical phases and time delays of ultrashort pulses in multiple non-collinear beams so that fully coherent four-wave and higher-order mixing measurements could be conducted without delay stages, multiple interferometers, or any active phase control. Coherences of biexcitons, unbound but correlated exciton pairs, and excitons undergoing rephasing were observed directly.

Keywords: ultrafast, multidimensional spectroscopy, many-body interactions, quantum well, pulse shaping

1. INTRODUCTION
Correlations between charged particles in semiconductors and other materials can produce bound electron-hole pairs known as excitons, whose wavefunctions are similar to those of hydrogen atoms. The charged particles can interact to form a hierarchy of electronic states which have fundamental importance and which may be used in applications such as quantum optics, quantum information technologies, and photovoltaic devices. In our system, the strength of the Coulomb force is large enough that multiple-particle interactions can influence the exciton dynamics via higher-order correlations. In fact, early optical four-wave-mixing (FWM) experiments on semiconductor quantum wells (QWs), showed that the frequency and dephasing rate of exciton coherences—such as the heavy-hole (H) and light-hole (L) excitons found in GaAs quantum wells—depended on the excited carrier density; the effects were dubbed excitation-induced shift (EIS) and excitation-induced dephasing (EID), respectively, and it was these types of effects that stimulated an intense interest in many-body correlations. Four-particle correlations, of which a bound biexciton is an example, have been especially intriguing because of their analogy to a hydrogen molecule and because the exciton and biexciton states represent correlations among successively greater numbers of electrons and holes. Measuring their associated coherences reveals information about complex many-body interactions and correlation dynamics. But until recently, spectroscopic investigations were indirect because two-quantum coherences involving four-particle correlations such as biexcitons do not radiate.

Here we use two-quantum, two-dimensional Fourier transform optical spectroscopy (2Q 2D FTOPT) to observe the dynamics of multixciton states directly. The approach is based on analogous methods in NMR, although in the optical regime maintaining the required phase relationships between beams is much more difficult than in the microwave or infrared spectral regimes. In 2Q 2D FTOPT, we use a spatiotemporal femtosecond pulse-shaping apparatus to overcome the challenge of controlling multiple, noncollinear, phase-coherent optical fields. The fields are in beams arranged in a BOXCAR geometry to measure the generated signal through wavevector matching in a background-free direction. Our results from a prototype GaAs quantum well system reveal several distinct two-quantum peaks. Coherences due to pure biexcitons (which consist of two identical excitons, H and H or L and L) are measured using a cross-linear polarization scheme. Coherences due to mixed biexcitons (which consist of two different excitons, H and L or L and H), are measured using a co-circular polarization scheme. In addition to coherences due to these bound states, we also observe coherences due to unbound but correlated exciton pairs, where the spins are unpaired. We can extract the biexciton binding energies and dephasing dynamics from peaks in the 2D spectra, and changes in the dephasing rates as a function of the excitation density are observed as linewidth changes. The dephasing rate variation suggests that still higher-order correlations due to exciton-biexciton interactions exist.

We can extend the measurements to a third dimension in 3D FTOPT by scanning both time periods and measuring the resulting spectrum. Since the full complex signal field is measured as a function of all three time intervals, nearly every
peak in the resulting 3D spectral solid arises from a distinguishable sequence of transitions represented by a single Feynman pathway. This fully coherent measurement allows unambiguous resolution of the coherent dynamics. We have used the 3D spectral peaks to separate two pathways involving mixed biexcitons generated in different time orders. In the process, we revealed a peak that was previously obscured by a correlated but unbound exciton pair coherence.

2. EXPERIMENTAL

The experimental apparatus—detailed previously^{3,4,11,15}—is mentioned here briefly. A Ti:sapphire oscillator was adjusted to create near-transform-limited 100 fs pulses centered at 806 nm. This wavelength corresponds to the energy of the H exciton resonance, 1540 meV. The apparatus, illustrated in Fig. 1, combines crossed static binary phase masks, whose four first-order diffracted beams are collected to create the BOXCARS geometry, with a 2D femtosecond spatiotemporal pulse shaper used in diffraction mode^{14} to control the field in each beam, including all temporal delays and phase shifts. The phase stability of the apparatus was measured to be $\lambda/88$ after eight hours. Diffraction-based pulse shaping permits independent control over the phase and amplitude profiles of the four outputs and eliminates the replica pulses that ordinarily accompany pulse shaping outputs. Eight-step phase cycling is used to reduce undesired signals due to scattered light from both the sample and the spatial light modulator (SLM) surface. The relative timing of the pulse or pulses are scanned, and the emitted signal is detected through spectral interferometry^{16} using a charge coupled device and spectrometer, with a typical data acquisition time of about 10 minutes for a 2D scan or 10 hours for a 3D scan. The radiated signal was spatially overlapped with a reference pulse, or local oscillator (LO), whose intensity is attenuated by $10^{-4}$ compared to the other pulses and whose arrival time at the sample is set to 1 ps before that of the final input field. The LO allows the full complex signal field to be acquired so that the real and imaginary surfaces can be extracted if desired. To control the polarization of each field, waveplates are placed in each beam before the final lens which focuses the beams into the sample (QW).

Figure 1. The FTOPT spectrometer. The input pulse is focused by a spherical lens (SL1) onto the phase mask (PM), whose first-order diffracted beams are collected by a second lens. The four identical beams in the rotated BOXCARS formation then pass through the beam splitter (BS) into the diffraction-mode spatiotemporal pulse shaper where they are dispersed by the grating (G) and collimated onto the four separate regions of the SLM surface by a cylindrical lens (CL). A pulse is delayed by imparting a linear phase sweep across its horizontally dispersed frequency components, as shown in the three regions $\phi_a$, $\phi_b$, and $\phi_{LO}$—that yield various pulse delays relative to the unshifted pulse, $\phi_c$. The phase of the carrier frequency, $\omega_0$, is held constant throughout the scan and is calibrated through additional measurements. The focus at the spatial filter (SF) is re-imaged onto the sample (QW) and the signal and LO are sent to a CCD spectrometer. The beams traverse a common set of optics, and the system is passively phase stable.
3. RESULTS

By varying the relative time-orderings, polarizations, and spectral content of each field, a large number of different scans are possible. Here we present three spectra showing how different many-body interactions can be isolated. In each case, the four beams were in the BOXCAR geometry, and the only difference between the scans was how the computer controlled the relative timings. Beam polarizations were controlled using manually adjusted waveplates.

3.1 Two-dimensional one-quantum rephasing spectroscopy

The spectrum shown in Figure 2 is the result of a scan in which the conjugate field—field $E_a$ in Figure 1—was scanned so that exciton coherences which oscillate during the scanned time period are rephased during the emission time. All of the fields had the same linear polarization, and the real part of the spectrum is plotted. This kind of scan was studied in detail elsewhere$^{8,10,17}$, and it is an instructive example of how many-body interactions manifest themselves in the peaks of a 2D spectrum. Unlike rephasing scans of samples without strong many-body interactions such as atomic vapors$^{18}$, here the peaks have dispersive lineshapes. This was shown to be due to the excitation-induced dephasing effect$^8$. A second many-body interaction is the presence of an excited-state absorption feature$^{19}$ which is red-shifted from the H exciton diagonal peak. This feature is due to biexciton-exciton emission, and the red-shift is due to the binding energy of the biexciton. A third striking feature of the spectrum is a vertical stripe on two of the features which absorbed at the L energy. This stripe can be modeled using excitation-induced dephasing.

![Figure 2. The real part of a rephasing (photon echo) scan measured using co-linearly polarized excitation and LO fields. Several many-body interactions manifest themselves in the peaks. The dispersive lineshape is due to excitation-induced dephasing. Biexciton-exciton emission is observed as a red-shifted shoulder to the H diagonal peak at about 1540 meV. The one-quantum diagonal ($E_{\text{abs}} = E_{\text{emit}}$) is the dashed line.](image)

3.2 Two-dimensional two-quantum nonrephasing spectroscopy

Two-quantum 2D FTOPT spectroscopy is used to separate the signal contributions due to four-particle correlations from signal contributions due to two-particle interactions$^3$. By creating and then measuring two-quantum coherences, biexciton properties can be directly measured. In Figure 3 we show a spectrum which allows us to extract the HH biexciton binding energy. In this two-quantum scan, the two nonconjugate fields—labeled $E_b$ and $E_c$ in Figure 1—were scanned as a pair. They created a biexciton–ground-state coherence that evolved during the two-quantum time period and that was projected to a one-quantum radiative coherence at a later time by the conjugate field, $E_a$. The coherent oscillations during the two-quantum time period were Fourier transformed to yield a 2D spectrum correlating the two-quantum coherence frequencies to the emitted signal frequencies. In this polarization configuration, fields $E_b$ and $E_c$ had cross-linear polarization and fields $E_a$ and $E_{\text{LO}}$ had cross-linear polarization; signal contributions due to excitation-
induced effects are eliminated. Thus, the main peak is well separated from other strong features, and the biexciton binding energy ($\Delta B$) can be extracted by measuring the shift of the peak below the two-quantum diagonal ($E_{2Q} = 2E_{\text{emit}}$).

Figure 3. Amplitude of a two-quantum 2D FTOPT scan. This spectrum was measured using a cross-linear polarization scheme to eliminate signal contributions due to excitation-induced effects. Only one main feature, which is due to HH biexciton–ground-state coherences, is observed. The spectrum of each excitation pulse was adjusted so that only H excitons were excited. We measure the binding energy of the HH biexciton ($\Delta B$) to be $1.0 \pm 0.1$ meV using the distance of the peak below the dashed two-quantum diagonal, $E_{2Q} = 2E_{\text{emit}}$.

3.3 Three-dimensional two-quantum nonrephasing spectroscopy

Often, we increase the dimensionality of a spectroscopic measurement when we desire to separate signal contributions which overlap. This was demonstrated long ago in NMR, was extended to the infrared, and only recently was it demonstrated in the visible. As shown in Figure 4, we can use 3D FTOPT to separate different pathways to mixed biexciton formation. Initial projection through the emission dimension onto the plane correlating initial absorption to two-quantum frequency shows several overlapping signals. By selectively integrating around the L exciton energy in the emission dimension and around the mixed biexciton energy in the two-quantum dimension, we can project onto the same plane, but now most of the signal contributions have been removed. The only two remaining signal contributions are due to two isolated pathways in which initial absorption was at H for $a_i$ and at L for $b_j$. After this initial event, both pathways contained mixed-biexciton–ground-state coherence during the two-quantum time period and then emitted as a L–ground-state coherence. Even though the number and type of interactions are the same, two events involving L and two events involving H, the peaks are of different intensities. Without the added power of 3D FTOPT, the signals would overlap in any two-dimensional measurement and this peak difference would not be measured.
The many-body interactions among excitons and associated several-particle correlations in quantum wells have been investigated for some time. Two-dimensional spectroscopy has helped isolate, measure, and understand how the different interactions manifest themselves in the sample. Here we presented three spectra, a 2D one-quantum rephasing spectrum, a 2D two-quantum nonrephasing spectrum, and a 3D two-quantum nonrephasing spectrum, which all showed clear many-body interactions. Two-quantum nonrephasing scans are useful for measuring the properties of four-particle correlations such as biexcitons. Future work will focus on extending the measurements to higher orders to reveal ever higher numbers of correlated particles.

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