Two-Quantum Coherences in Optical Two-Dimensional Fourier Transform Spectroscopy

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INTRODUCTION. Multidimensional Fourier transform spectroscopy is a powerful tool for studying the coherent optical response of complex systems. By tracking the phase of a nonlinear signal as a function of the delays between the excitation pulses, coupling between resonances can be determined. Moreover, the evolution of coherences that do not have a dipole associated with them can be observed through the phase they impart on the signal. One example of this is a two-quantum coherence between the ground state and a doubly excited state. Two-quantum coherences have been observed in IR two-dimensional Fourier transform (2DFT) spectroscopy of molecular vibrations [1,2].

Here, we present the results of experiments using an appropriate pulse sequence to probe two-quantum coherences of electronic excitations in semiconductors and a potassium vapor. The results show that resonant two-quantum coherences do occur even though in a single particle picture there is not a doubly excited state resonant with the laser. In semiconductors, this result is due to the strong many-body interactions. In the atomic vapor, simulations show that the signals arise due to atom-atom interactions.

SEMICONDUCTOR EXCITONS. Two-quantum coherences are expected to occur in semiconductors between the ground state and the biexciton state, and have been observed [3]. By spectrally selecting the heavy-hole (HX) exciton and measuring the real part of the 2DFT spectrum, Fig. 1(a), we observed that there are signals at the two-quantum frequency of the biexcitons, but also at the frequency of twice the exciton, i.e., not red-shifted by the biexciton binding energy [4,5,6]. In addition, the two-exciton signal appears for co-circularly polarized excitation, which cannot create a bound biexciton from two heavy-hole excitons. Simulations show that these signals arise from mean-field many-body interactions [6], in agreement with the dispersive linshape.

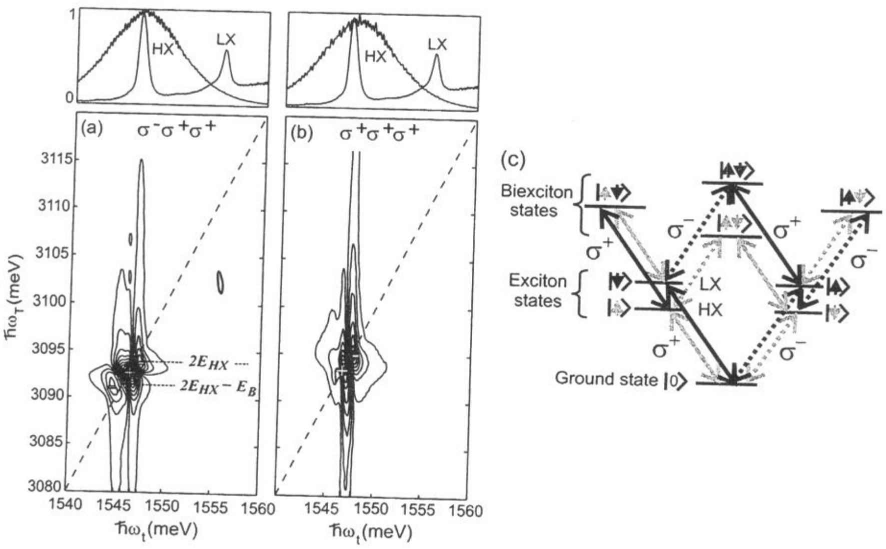

Fig 1. 2D two-quantum spectra of GaAs quantum wells at different polarizations. (a) Experimental spectra obtained using cross-circular ($\sigma^-\sigma^+\sigma^+$)) and (b) using co-circular ($\sigma^+\sigma^+\sigma^+$) polarization. (Top) The laser excitation is resonant with HX energy. EHX is the heavy-hole exciton energy, and EB is the biexciton binding energy. (c) Level scheme for the heavy- and light-hole excitons and biexcitons in GaAs quantum wells.

POTASSIUM VAPOR. As a model system, we have studied the 2DFT spectra of the D_1 and D_2 lines in a potassium vapor. There is good agreement between experiment and theory for one-quantum spectra [7]. Figure 2 shows the 2-quantum 2DFT spectrum of a potassium vapor around the D_1 and D_2 lines. The level structure of an isolated potassium atom is shown at right. Within the bandwidth of the laser, there are no levels at the energy of twice the D_1 and D_2 transitions, thus it should naively be expected that there would be no 2-quantum signals. Nevertheless strong signals are observed.

The observed 2-quantum coherences in a potassium vapor is due to interaction between atoms. In a simple picture, a pair of two-level atoms can be viewed as a four level system, with a ground state, two singly excited states and a doubly excited state. The two-quantum coherence is between the ground state and the doubly excited state. If the atoms do not interact, signals from final coherences between the ground state and singly excited state exactly cancel those between the singly and doubly excited states (in both case the two quantum coherence is an intermediate state). However, if the two atoms interact, then the cancelation does not occur and a signal will be observed. The experimental results are reproduced by a model with dipole-dipole interactions between atoms that move on trajectories determined by a molecular dynamics simulation.

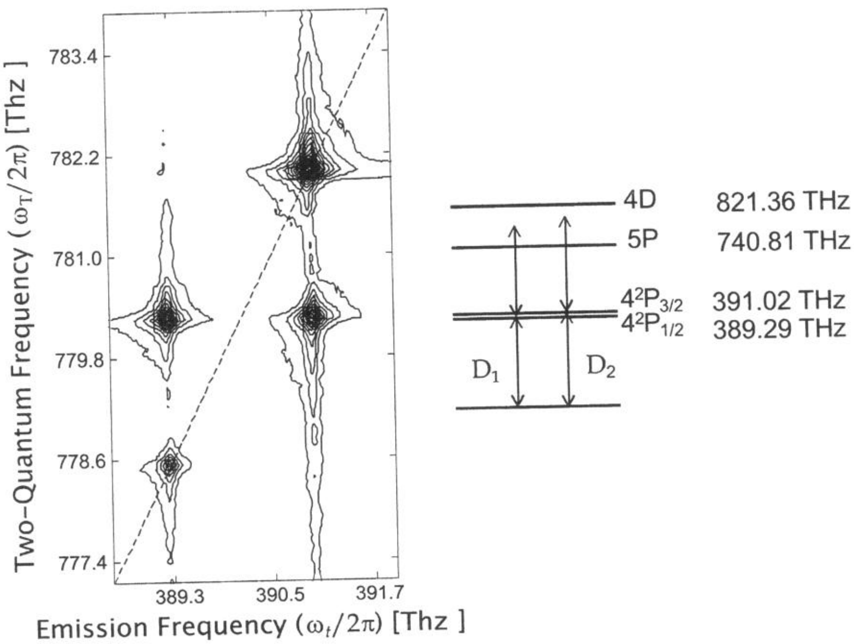

Fig 2. Amplitude two-quantum spectrum of a potassium vapor. The level structure of an isolated potassium atom is shown at right.

SUMMARY. We have shown two-quantum 2DFT spectra of excitons in semiconductors and atoms in a vapor. In both cases, the two-quantum spectra are very sensitive to interactions, specifically many-body interactions in semiconductors and dipole-dipole interactions between atoms in a vapor.

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