Polarization-dependent absolute-phase-corrected multidimensional coherent spectra of exciton-polaritons

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ABSTRACT

Multidimensional coherent spectroscopy measures the third-order polarization response of a system to reveal microscopic electronic and many-body phenomena. Applied to semiconductor nanostructures, it can distinguish homogeneous and inhomogeneous broadening due to disorder or strain gradients, resolve coupling between transitions, and optically access transitions that are either non-radiating or outside the bandwidth of the pulses. Two tools often exploited in this versatile technique are (i) the ability to control the polarization of the excitation and emission and thus the optical selection rules, and (ii) the ability to capture the complex spectrum. Here, the polarization of pulses emerging from a multidimensional optical nonlinear spectrometer (MONSTR) and the resulting four-wave mixing emission are controlled automatically using variable retarders, such that multiple spectra are recorded during a single phase-stabilized scan. This improves the acquisition time by $\sim 3\times$ compared to running separate polarization scans. Importantly, only one phase ambiguity exists in the complex spectra across all sets of polarization states measured. This single ambiguity is resolved by comparing the initial spectrally resolved transient absorption to the complex four-wave mixing spectrum for collinear polarization and then applying it to all spectra. Here, the method is applied to a quantum well embedded in a semiconductor microcavity with an adjustable cavity-exciton detuning. The complex 2DCS spectra we report constitute the first measurements of detuning- and polarization-dependent exciton-polariton lineshape across the strong coupling regime.

Keywords: Multidimensional coherent spectroscopy, Coherent coupling of Exciton-Polariton, Phase corrected 2D Spectra, Four wave mixing

1. INTRODUCTION

Optical properties of semiconductor quantum wells (QW) are dominated by excitonic resonances at low temperature. Excitons are Coulomb bound electron-hole pairs. When such a QW is placed inside a planar semiconductor microcavity at the antinode of the resonant field of the cavity, the QW exciton strongly interacts with the cavity normal mode, resulting in the formation of a new quasiparticle, exciton-polaritons.[1] The single exciton line is replaced with lower polariton (LP) and upper polariton (UP) branches, with a splitting characterized by the vacuum Rabi splitting energy E_{VRS}. Exciton-polaritons are hybrid modes composed of an exciton-like part and a light-like cavity photon mode part. The energies of the LP and UP branches and how these parts are divided between them depends on the detuning (Δ) of the cavity mode with respect to the exciton energy. The detuning-dependent branch energies are depicted in Fig. 1(a). The exciton and photon fractions of the UP or LP branches are represented by Hopfield coefficients,[2] H_x and H_{cav}, respectively, which are shown in Fig. 1(b) as a function of the cavity detuning. In the present study, we employed two-dimensional coherent spectroscopy to investigate exciton-cavity coupling over a range of detuning values near the zero detuning of the cavity.

Two dimensional coherent spectroscopy (2DCS) is an enhanced version of transient four wave mixing (TFWM) which has been developed over the last two decades into an important tool for studying ultrafast coherent processes in semiconductor nanostructures.[3–8] Multidimensional spectroscopy has numerous advantages over linear

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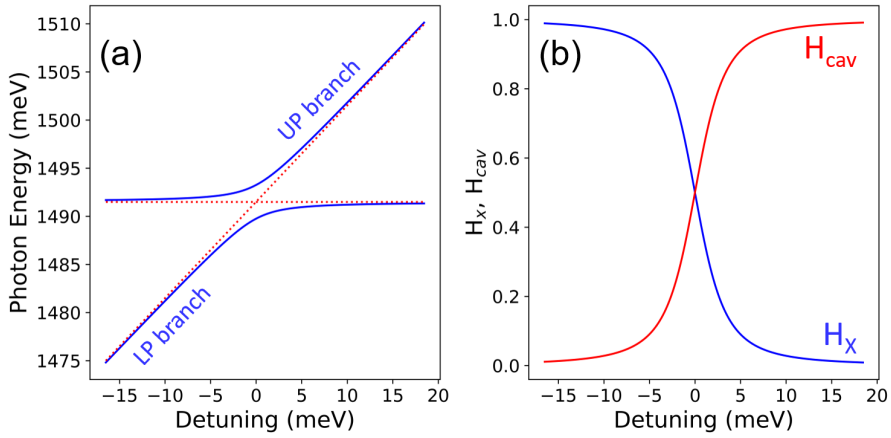

Figure 1. (a) Illustration of anticrossing of LP and UP branches in the semiconductor microcavity studied. The branch energies are shown as blue solid lines. The red dotted lines show the bare exciton (horizontal) and cavity mode (linear slope) energies in the absence of interaction. The splitting at zero detuning is the vacuum Rabi splitting E_{VRS}. (b) Hopfield coefficients of the UP branch plotted as a function of the cavity detuning. H_x and H_{cav} represent the exciton and photon fractions of the branch, such that $H_x + H_{cav} = 1$. H_x and H_{cav} are reversed for the LP branch.

spectroscopy techniques or conventional one dimensional time domain spectroscopy techniques as it correlates two time domains and represents the correlation spectra in a two dimensional map in the frequency domain. In a 2D spectrum, the resonant transition frequencies appear on the diagonal and any coupling between the resonances are revealed by the cross diagonal peaks. Another advantage of 2DCS is that homogeneous broadening can be easily isolated from inhomogeneous broadening a rephasing 2DCS scan. The homogeneous and inhomogeneous linewidths of a resonance can be directly obtained by finding cross-diagonal and diagonal slices of the resonance lineshape,[9] respectively, in a rephasing 2D spectrum. Polarization dependent 2DCS measurements isolate various quantum pathways and allow identification of many body interactions that contribute to the third order nonlinear response of the material.[10] Proper phasing of the 2D spectra allows extracting the signal phase. Previous work on quantum wells in microcavities reported magnitude 2D spectra, not fully phased spectra.[11–14]

In this proceeding, we present polarization-dependent complex 2D spectra, phased using spectrally-resolved transient absorption (SRTA) measurements. Real spectra of rephasing 2DCS for a range of cavity detuning values are measured as well as the polarization dependence for a single (near-zero) detuning. To our knowledge, this is the first report of phased 2DCS in microcavity samples and is the first step to understanding the full $\chi^{(3)}$ response of this strongly nonlinear optical system.

2. EXPERIMENTAL DETAILS

The experimental setup used in our study is well documented in Ref. 15 and will be only briefly described here. A 120 fs mode locked laser pulse from a Ti:sapphire oscillator with a repetition rate of 76 MHz is used as an input to a Multidimensional Optical Nonlinear SpecTrometeR (MONSTR)[16] instrument to generate four identical phase stabilized pulses. In Fig. 2(a), the pulse sequence for a rephasing scan is shown, where the phase conjugated pulse A* and pulse B are separated by time delay τ, and pulse B and pulse C are separated by time delay T. The beams are arranged in a box geometry, i.e. on the corner of a square, as shown in Fig. 2(b). The three excitation pulses, separated by two time delays τ and T, are focused on the sample mounted inside a liquid He cooled cryostat using a single lens and the resulting four-wave mixing (FWM) signal is emitted in the phase matching direction (along the fourth pulse). In a rephasing pulse sequence, the phase conjugated pulse arrives first on the sample followed by the other two pulses, and the FWM signal is emitted as a photon echo in the phase matched direction ($-k_A + k_B + k_C$, where k_i is the wavevector of excitation pulse i). The FWM signal is then collinearly combined with a phase stabilized local oscillator and the resulting spectral interferogram is dispersed into a grating spectrometer, and is then detected by a thermoelectrically cooled CCD. In this work,

rephasing 2D spectra were measured for four different polarization configurations and the time delay T is set to ≈ 50 fs. The polarizations of the incident laser pulses were controlled by liquid crystal variable retarders. In order to reduce artifacts caused by scattered light in the direction of the FWM signal, the phase of pump pulses A and B were phase cycled using two additional liquid crystal variable retarders. Phase cycling also improves the signal to noise ratio.

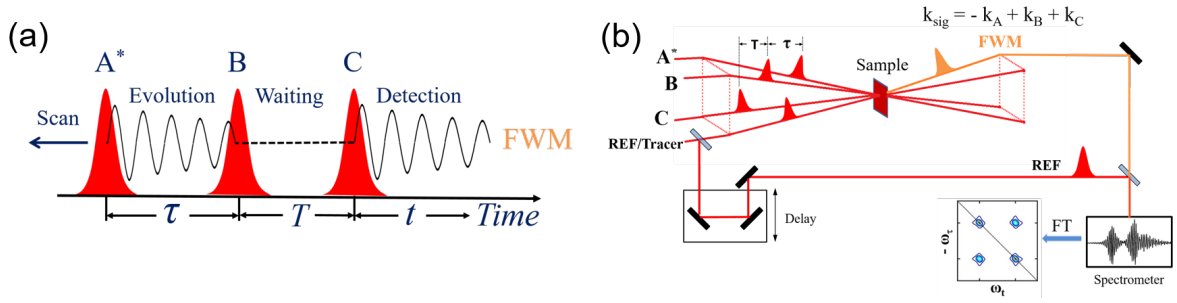

Figure 2. (a) Pulse sequence for a rephasing scan. (b) Pulse arrangement in the box geometry used in the experiment. The four-wave mixing (FWM) beam is emitted in a background-free, phase matching direction $-k_A + k_B + k_C$. The local oscillator (REF) is sent around the sample.

The microcavity sample used in our study was grown by molecular beam epitaxy on a GaAs substrate. The mirrors consist of GaAs/AlAs (14.5 and 12 bilayer) distributed Bragg reflectors separated by a wedged λ GaAs cavity, with a cavity mode close to 830 nm (1491 meV), the energy of the heavy hole exciton. A single 8 nm $In_{0.4}Ga_{0.96}As$ QW is placed at the antinode of the cavity.[1,12] The sample has a vacuum Rabi splitting parameter $E_{\mathrm{VRS}} = 4$ meV. The cavity is slightly wedged, which allows us to vary the detuning by translating the sample in a direction perpendicular to the incident laser beams. Polaritons are very sensitive to the angle of incidence. In our experiment, the three excitation laser beams were incident on the sample at an external angle of approximately 7° to the normal.

Because of the strong interaction of the microcavity with the light, the optical density and dispersion of the sample is much higher than a typical semiconductor sample. As a result, the all-optical phasing technique often used with the MONSTR did not reliably work. Instead, we measure the spectrally-resolved transient absorption (SRTA) spectrum for the VVVV polarization configuration at the beginning of each scan. This spectrum is the same as a slice of the phased 2D spectrum, which allows us to fit it to find the global phase for that polarization.[17,18] Because we use liquid crystal variable retarders to polarize the beams and capture data for each polarization configuration on every step in the 2D scan, we can apply the known polarization-dependent phase imparted on the beams, which allows us to set the global phase of each polarization configuration measured.[15] Phased complex two-dimensional coherent spectra (2DCS) for common polarization configurations were recorded for a GaAs multiple quantum well sample for comparison to previous work.[10,15]

3. RESULTS AND DISCUSSION

Magnitude-only 2D spectra for this microcavity sample were presented in Refs. 12 and 14. In Ref. 15, preliminary phased polarization-dependent 2D spectra were presented for $\Delta = -6.0$ meV, where the lower branch is primarily light-like and the upper branch is primarily exciton-like. It was found that the diagonal peak corresponding to the exciton-like branch had the same dispersive lineshape as seen for the heavy hole exciton in a bare quantum well sample.[10] However, the sign of the biexciton feature was opposite what is seen in a bare quantum well sample. Here we present phased 2D spectra for values of detuning nearer the avoided crossing ($\Delta \approx 0$) and show how the phased 2D spectrum evolves with Δ.

Figure 3 depicts the rephasing amplitude (a-d) and real part (e-h) of the 2D spectrum for four different polarization configurations at near zero detuning ($\Delta \approx 0.05$ meV). Here, VVVV, VHHV, $\sigma^+\sigma^+\sigma^+\sigma^+$, and $\sigma^-\sigma^-\sigma^+\sigma^+$ represent the co-linear, cross-linear, co-circular, and cross-circular polarization configurations, respectively with first three letters representing the polarizations of the first three pulses, while the fourth letter corresponds to

the emission polarization. The lower and upper polariton branch energies on the diagonal are labeled as LP and UP, respectively. Cross-peaks corresponding to couplings between the branches are labeled as UP-LP and LP-UP.

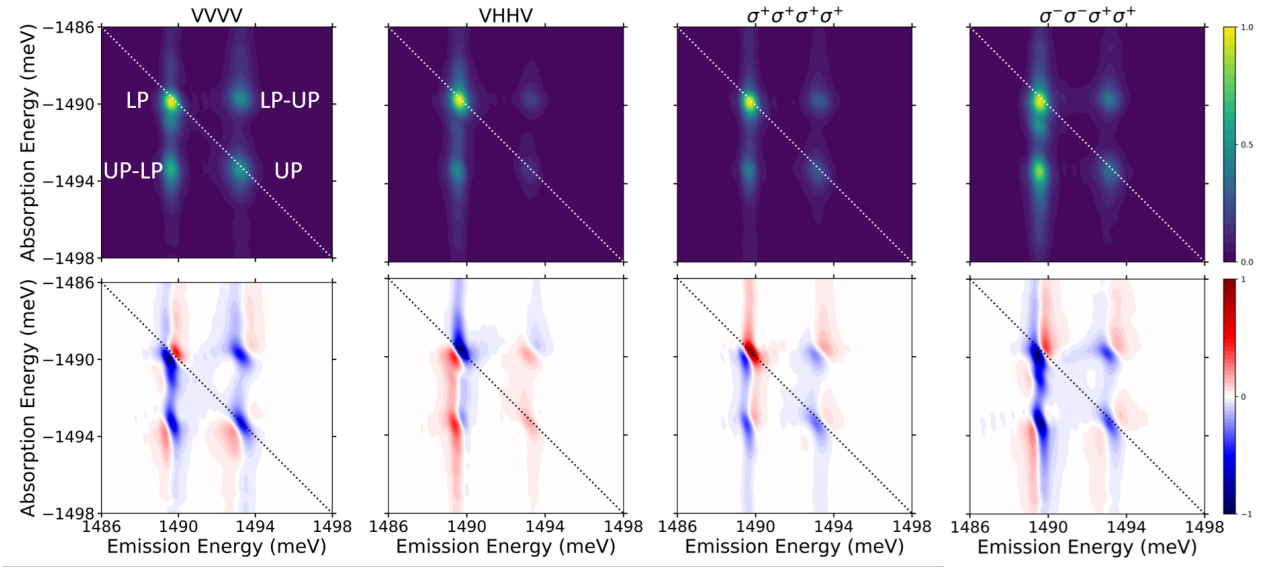

Figure 3. Polarization dependent rephasing 2D Spectra at near zero detuning, $\Delta \approx 0.05$ (meV). (a)-(h) The magnitude and the real part of the complex 2D spectra are shown in the top and bottom rows, respectively. VVVV, VHHV, $\sigma^+\sigma^+\sigma^+\sigma^+$, and $\sigma^-\sigma^-\sigma^+\sigma^+$ represent the co-linear, cross-linear, co-circular, and cross-circular polarization configurations, respectively.

The detuning dependence of the amplitude spectra we measure are consistent with previous results on this sample,[12] so we focus here on the real part spectra. Fig. 4 shows the real part of the rephasing spectrum for Δ values ranging from -3.27 meV to 3.37 meV.

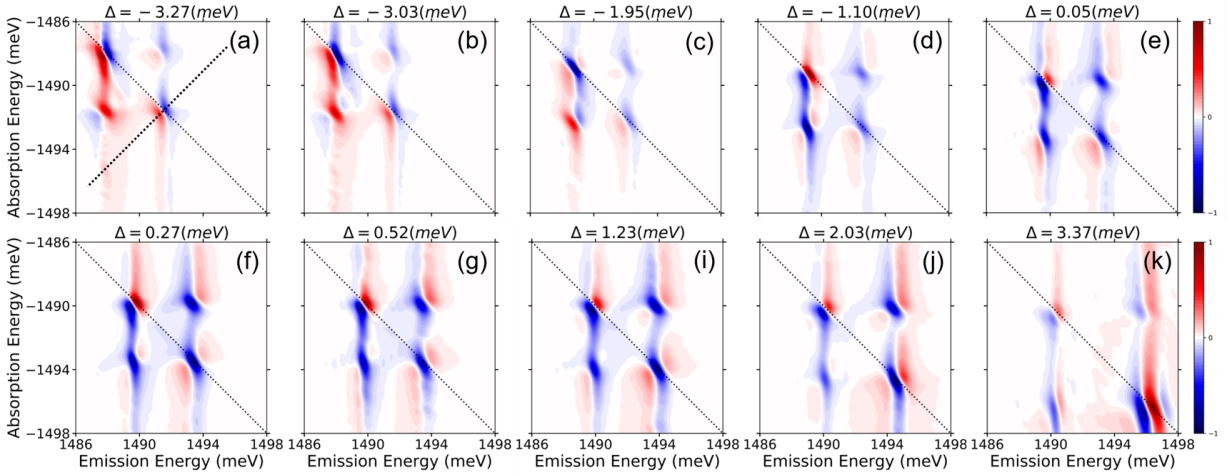

Figure 4. (a)-(j) Real Part of the rephasing 2D Spectra for different detuning values (Δ) in the range of -3.27 meV to 3.37 meV, (shown on top of each spectrum), for co-linear polarization configuration. Each spectrum is normalized to the maximum of the two diagonal resonances.

We find that the phase of the spectrum evolves approximately smoothly as we change the detuning parameter, as expected. This can be seen most clearly in Fig. 5, which shows line outs of the 2D spectra as a function of Δ.

A detailed analysis of the phased spectra and comparison to theoretical models will be explored in future work.

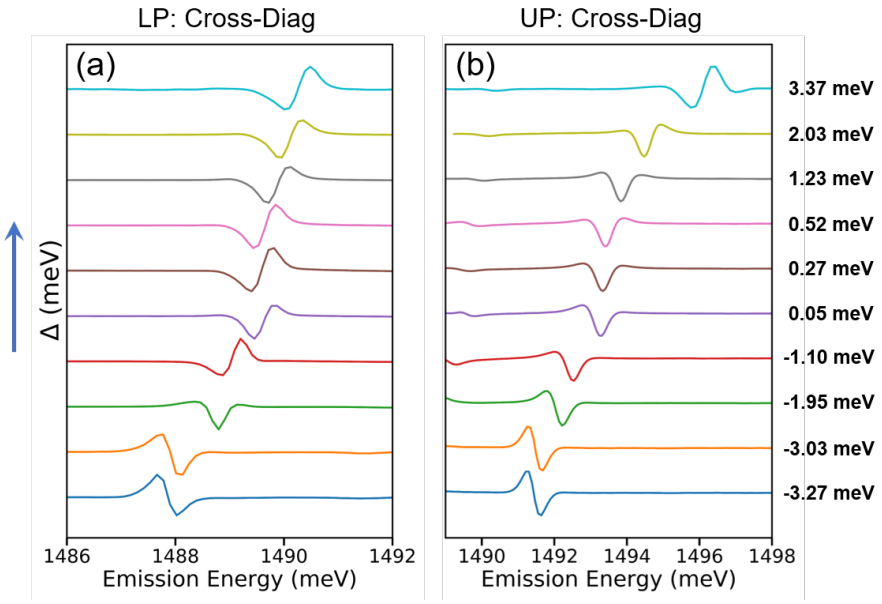

Figure 5. Cross-diagonal profile of real part 2D spectra of LP and UP as a function of detuning.

4. CONCLUSION

In summary, we have measured the response of a microcavity using multidimensional optical coherent spectroscopy. We investigate the coherent coupling between the exciton-like and cavity like normal modes in a semiconductor microcavity as a function of the detuning of the cavity. A full set of polarization dependent 2D spectra with co-linear, cross-linear, co-circular, and cross-circular polarization configuration were collected and phased with the help of spectrally-resolved transient absorption (SRTA) measurements. We observe the phase of the upper and lower polariton branches evolve gradually as a function of detuning.

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