Rabi Oscillations in an InAs Quantum Dot Ensemble Observed in pre-pulse 2D Coherent Spectroscopy

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Abstract: Pre-pulse 2D coherent spectroscopy is used to observe Rabi oscillations in an InAs quantum dot ensemble. The 2D spectra can monitor the coherent evolution in an ensemble system despite inhomogeneous broadening.

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A semiconductor quantum dot (QD) is one of the most promising candidates for use as a qubit in quantum information devices as the large dipole moments μ and the discrete level system in QD enable us to control the light-matter interaction coherently. Optical Rabi oscillations are a fundamental demonstration of coherent manipulation, which manifest themselves as the sinusoidal dependence of the population inversion in a two level system on the pulse area $\int_{-\infty}^{\infty} dt\mu E(t)/\hbar$ ($E(t)$ is a envelope function of the driving electric field). Compared to the demonstrations of coherent control for single QDs [1], there have been few studies for ensemble QDs [2,3], which is essential for the realization of large scale quantum devices using multiple qubits. 2D coherent spectroscopy (2DCS) has the advantage of mapping signals into two dimensions, which enables us to obtain information hidden in one dimensional spectroscopy, such as couplings between multiple states or clear separation between inhomogeneous and homogeneous broadening [4]. This advantage also serves as a tool to clearly distinguish between contributions from a lower state and an upper state in an optically excited QD ensemble, which is discussed later.

In this work, we have investigated Rabi oscillations in an InAs QD ensemble by using pre-pulse 2DCS, which is a pump-probe experiment, where the pre-pulse is the pump and the 2DCS works as the probe (Fig. 1 (a)). 2DCS is a three-pulse transient four wave mixing (TFWM) technique with the addition of interferometric stabilization of the pulse delays. Three pulses are incident on the sample in a rephasing (photon echo) time-ordering and the TFWM signal is spectrally-resolved while the delay between the first two pulses is varied. A Fourier transform of the signal with respect to this delay generates a 2D spectrum, in which the excitation and emission energies are correlated along the vertical and horizontal axes, respectively.

The sample investigated is a self-assembled InAs/GaAs QD ensemble, consisting of 10 quantum-mechanically-isolated epitaxially-grown layers that are thermally annealed post-growth at 900°C for 30 s, which results in a 100 meV in-plane confinement, i.e. a blue-shift due to Ga-diffusion into the QDs, which is beneficial for detecting the signals with Si avalanche diodes [5]. The sample was unintentionally doped during growth, resulting in approximately

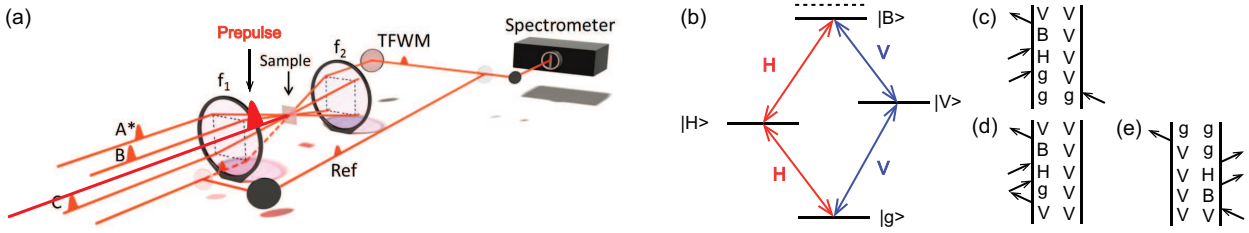

Fig. 1: (a) Schematics in pre-pulse 2DCS experiment. (b) The energy level diagram showing the horizontal (H) and vertical (V) exciton and biexciton (B). The quantum path for VHHV polarization starting with population in the ground state (c) and the V exciton state ((d) and (e))

half of the QDs being charged with a hole. The lowest exiton transition is four-fold degenerate with two optically-active (bright) states that are coupled through the exchange interaction, forming an eigenbasis of the horizontally (H) and vertically (V) polarized states for asymmetrically shaped QDs (Fig. 1 (b)). The sample is aligned such that the eigenstates are along the H and V directions. All of the measurements are performed with the sample cooled at 10 K in a sample in vapor flow cryostat.

A cross-linearly polarized excitation and detection scheme is used, i. e. VHHV for A, B, C and the TFWM signal respectively, which isolates the biexciton signals from trions. Figure 2 (a) shows a typical 2D rephasing amplitude spectrum without pre-pulse. The spectrum features two peaks that are inhomogeneously broadened along the diagonal direction due to QD size dispersion. The peak labeled D arises from the trion nonlinear response, whereas the peak labeled B1 arises from the biexciton nonlinear response and is red-shifted along emission energy by the biexciton binding energy corresponding to the quantum path shown in Fig. 1 (c). The polarization for the pre-pulse is set along the V direction, which creates the superposition between the ground state and the V exciton state, and other quantum paths starting with population in the V exciton state (Fig. 1 (d) and (e)) contribute to the 2D signal. Figure 2 (b) shows a 2D rephasing amplitude spectrum at a prepulse intensity of 8.8 μJ/cm^2. Compared to Fig. 2 (a), the B1 signal becomes weaker, due to the quantum paths shown in Fig. 1 (d), which has the opposite sign to the signal shown in Fig. 1 (c). Additionally, a peak labeled B2 arises from the biexciton nonlinear response from the V exciton state corresponding to Fig. 1 (e) and is blue-shifted along emission energy by the biexciton binding energy. Using 2DCS, the biexciton nonlinear responses starting with population in the ground state and the V exciton state can be clearly separated, enabling us to estimate the ratio of the population in the V exciton state to the ground state quantitatively in an ensemble QD. Figure 2 (c) and (d) show the amplitude of peaks B1 and B2 in red circles and blue squares, respectively, as a function of pulse area which is proportional to the square root of pre-pulse intensity. B1 gets weaker, while B2 gets stronger with increasing pulse area. The solid red line in Fig. 2 (c) is a fit to a damped oscillator model given by $A\exp(-\gamma\sqrt{I})\cos(a\sqrt{I})$, where γ is a decay of coherence, I is pre-pulse intensity, and A and a are the scaling parameters. From the fitting, we estimate that a pulse area of up to 0.41 π is achieved in our experiment. Work on increasing the pulse area to study highly excited coherent state is on going.

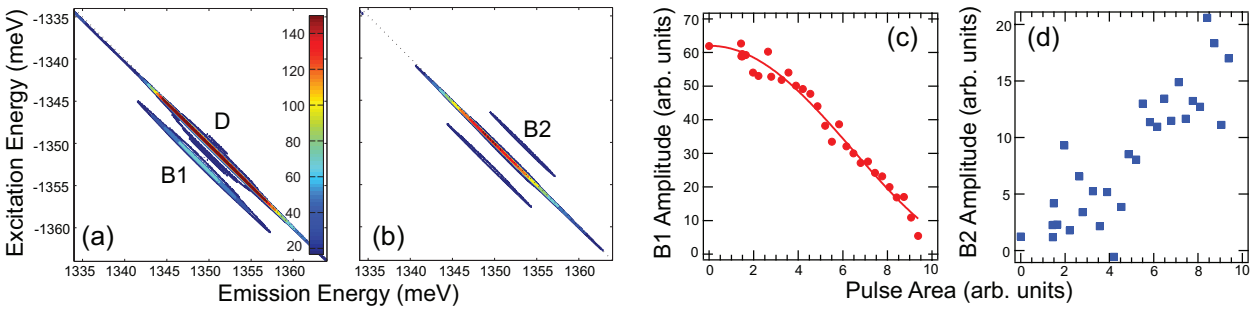

Fig. 2: 2D rephasing amplitude spectrum for an InAs self-assembled QD ensemble at prepulse intensity of (a) 0 and (b) 8.8 μJ/cm^2. Color scales in both (a) and (b) are shown at the colorbar in (a). Amplitude for peak for B1 (c) and B2 (d) as a function of electric field which is proportional to the square root of pre-pulse intensities.

In summary, we have observed Rabi oscillations in an InAs QD ensemble using pre-pulse 2DCS. The 2D spectra can clearly separate the upper state's signal from the lower state, enabling us to monitor the coherent evolution despite the inhomogeneous broadening.

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