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A new method for coherent control in a quantum dot ensemble

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Abstract. We propose a new method for controlling macroscopic coherent phenomena by taking advantage of the characteristics of quantum dot (QD) ensembles. We demonstrate that the ensemble average of Rabi oscillations of excitonic polarization can be changed drastically by selecting the spatial distributions of input electric fields. We show that the changes in ensemble averages are caused by polarization interference; the results of our previous experiments agree with our calculations. This result demonstrates that a macroscopic coherent response from a QD ensemble can be controlled by changing the spatial distribution of excitation beams.

1. Introduction

Optical Rabi oscillations (ROs) play crucial roles in coherent control of excitons in semiconductor quantum dots (QDs). Most reported ROs of QD excitons were observed in a single QD [1-3] because a large inhomogeneous distribution of Rabi frequencies results in smearing out ROs in a QD ensemble. Thus, QD ensembles have been considered unsuitable for coherent manipulation.

In a previous study, we demonstrated an “ensemble effect” on the ROs of excitonic polarization and population by measuring four-wave mixing signals and differential transmission. The ensemble effect is defined as the pulse area distribution for each QD with an exciton; this distribution is attributed to the inhomogeneous distribution of exciton transition dipole moments and the spatial distribution of input electric fields [4]. The ensemble effect mainly causes strong damping of ROs. However, the effects on ROs of excitonic polarization are different from those on ROs of excitonic population. We first showed that part of the ensemble effect is cancelled out only in the ROs of excitonic polarization. This is explained well by polarization interference.

In this study, we propose a more positive way to control macroscopic coherent phenomena by taking advantage of the characteristics of QD ensembles. We demonstrate that the ensemble average of ROs of excitonic polarization can be changed drastically by selecting the spatial distributions of input electric fields. We show that the changes in ensemble averages are caused by polarization interference.

2. Calculation

2.1. Calculation Models

We assume that an excitonic system is a two-level system. According to the optical Bloch equations, in an ideal two-level system, the Bloch vector rotates from the lower to the upper state with an increase in the pulse area, as shown in Fig. 1 (a). In the case of excitonic ROs, the upper state is equivalent to the exciton ground state, and the lower state is equivalent to the crystal ground state, where no exciton exists in a QD. The excitonic polarization corresponding to the value of $\boldsymbol{v}$ is proportional to $\sin\Theta_1$, while the excitonic population corresponding to the value of $\boldsymbol{w}$ is proportional to $\sin^2(\Theta_1/2)$, where Θ_1 is the pulse area of pulse 1. By changing Θ_1, we expect to observe ROs of excitonic polarization and population consistent with the abovementioned equations. Two-pulse four-wave mixing (FWM) and differential transmission (DT) techniques were used to investigate the ROs of excitonic polarization and population, respectively. This is because that the amplitude of the FWM signals observed in the arrangement shown in Fig. 1 (b) is described by $\boldsymbol{P}_{\mathrm{FWM}} \propto \sin\Theta_1$ [5], while the DT observed in the arrangement shown in Fig. 1 (c) is described by $\mathrm{DT} \propto \sin^2(\Theta_1/2)$.

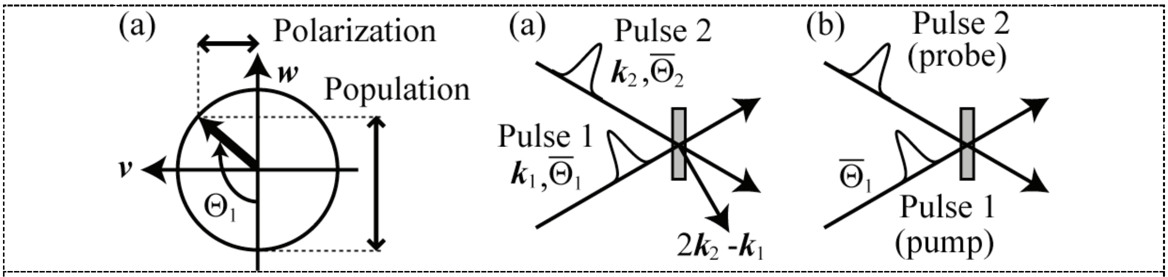

Figure 1. (a) Bloch sphere for a two-level system. Excitonic population is proportional to $\sin^2(\Theta_1/2)$, and excitonic polarization is proportional to $\sin\Theta_1$. (b) Experimental arrangement of FWM technique. (c) Experimental arrangement of pump-probe technique.

We calculated the ensemble average of FWM signals and DT, taking into accounts the spatial distributions of input pulse areas. The average pulse area $\overline{\Theta}_i$ of the $\boldsymbol{k_i}$ pulse is changed by varying the spatially averaged electric field $\overline{E}_i$ according to the equation $\overline{\Theta}_i = \overline{\mu}\,\overline{E}_i \Delta T/\hbar$, while maintaining a constant temporal duration ΔT for the a rectangular pulse. Here, we leave the dephasing process out of consideration. $\overline{\mu}$ and $\overline{E}_i$ represent the average transition dipole moment of the excitons and the average electric field, respectively. If the focused beam has a Gaussian spatial distribution, the QDs in the beam spot are excited not by the same electric field strength $\overline{E}_i$ but by different field strengths depending on their positions $E_i(x, y)$. This results in a spatial distribution of the pulse area for each QD at each position $\Theta_i(x, y)$. In this study, we consider that the spectral widths of the incident pulses are much smaller than the inhomogeneous broadening of the excitonic energies in QDs. The transition dipole moment μ is thus assumed to be homogeneous compared to $E_i(x, y)$.

Figure 2 shows the distribution functions of the excitation beam used in our calculations, i.e., (a) a flat-top beam, (b) a Gaussian TEM00 beam, and (c) a Gaussian TEM10 beam. The average electric field $\overline{E}_i$ is defined as $\iint E_i(x, y)dxdy/\pi R^2$, where R is the beam radius. Since it's reasonable to assume that R (larger than several μm) is much greater than lateral QD size and the distance between QDs (~typically several tens nm), we consider that a large number of QDs are excited resonantly in a beam spot.

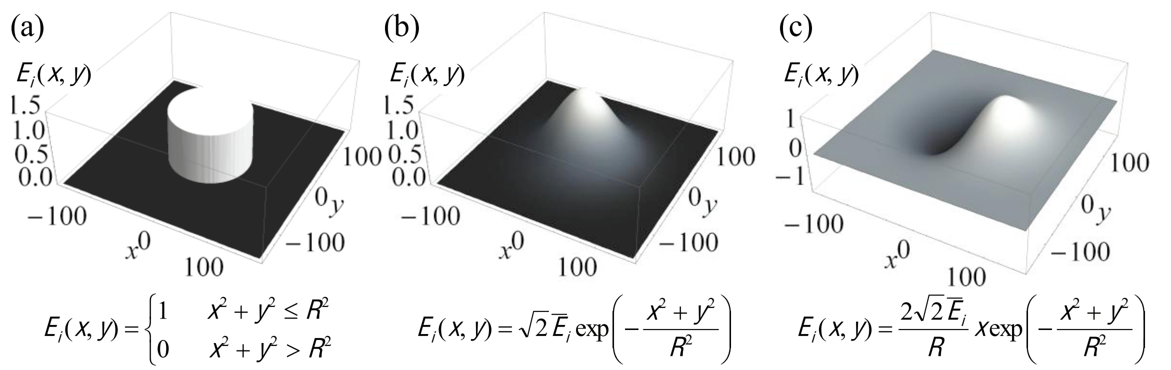

Figure 2. Spatial distribution of incident electric fields: (a) a flat-top beam, (b) a Gaussian TEM00 beam, and (c) a Gaussian TEM10 beam.

2.2. Calculation Results

Figure 3 shows the calculated FWM signals (a) and DT (b) as a function of $\overline{\Theta}_1$. Note that the horizontal axis shows the "average" pulse area $\overline{\Theta}_1$; the pulse areas for different QDs are distributed even at the same $\overline{\Theta}_1$ according to the distribution function of the electric field shown in Fig. 2. The spatial distribution of pulse 2 is assumed to be the same as that of pulse 1. $\overline{\Theta}_2$ was fixed at π in this study. As shown in Fig. 3(a), the ensemble average of ROs of excitonic polarization can be changed drastically by selecting the spatial distributions of input electric fields. The FWM signals excited by flat-top beams coincide with ROs in an ideal two-level system represented as $\sin\overline{\Theta}_1$. This is because the inhomogeneity of transition dipole moment μ is assumed to be negligible compared to the distribution of $E_i(x,y)$ in this study. In practice, the inhomogeneity of transition dipole moment slightly influences the RO; in other words, the ensemble of ROs excited by a flat-top beam will provide direct information on the inhomogeneity of μ. The FWM excited by Gaussian TEM00 beams coincides with $\sin\overline{\Theta}_1$ at $\overline{\Theta}_1 <\pi$ and shows the second peak at around $\overline{\Theta}_1 =1.5\pi$ although the amplitude is strongly damped. This is because of interference between excitonic polarizations with various amplitudes and directions [4]. The FWM excited by Gaussian TEM10 beams vanishes because negative and positive polarizations cancel out completely, which is much different from the calculated result for a TEM00 beam. As shown in Fig. 3(b), the behavior of DT is quite different from that of FWM signal. Though DT excited by flat-top beams coincides with ROs in an ideal two-level system represented as $\sin^2\left(\overline{\Theta}_1/2\right)$, DT excited by Gaussian TEM00 does not decrease with an increase in $\overline{\Theta}_1$ and there are few differences in DT between TEM00 and TEM10 beams. The reasons for this difference between FWM signals and DT are as follows: since the integration in the FWM signals involves the superposition of $\sin\overline{\Theta}_1$, which have both positive and negative values, the distribution of pulse area is partly cancelled out in the integration. On the other hand, $\sin^2\left(\overline{\Theta}_1/2\right)$ involved in DT have always positive values and the integration just smears out oscillation.

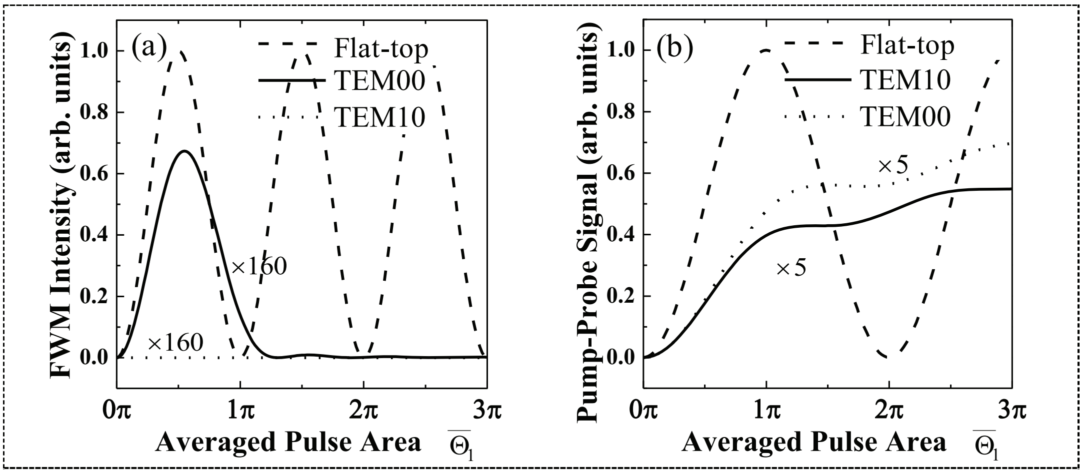

Figure 3. Calculations of Rabi oscillations of excitonic polarization (a) and population (b) excited by a flat-top beam (dashed line), a Gaussian TEM00 beam (solid line), and a Gaussian TEM10 beam (dot line).

3. Conclusion

We demonstrated that the ensemble average of ROs of excitonic polarization can be changed drastically by selecting the spatial distributions of input pulses due to the polarization interference. The effects of the spatial distributions are different between ROs of excitonic polarization and ROs of excitonic population. These results demonstrate that a macroscopic coherent response from a QD ensemble can be controlled by changing the spatial distribution of excitation beams.

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