Coherent control in inhomogeneously broadened quantum dots ensemble and its coherent transient phenomena

Noriaki Tsurumachi\(^{1,2}\), Kazuhiro Komori\(^{1,2}\), and Toshiaki Hattori\(^3\)

\(^1\)National Institute of Advanced Industrial Science and Technology (AIST), 1-1-4 Umezono, Tsukuba, Ibaraki, Japan
\(^2\)CREST—Japan Science and Technology Corporation (JST)
\(^3\)Institute of Applied Physics, University of Tsukuba, 1-1-1 Tennodai, Tsukuba, Japan

Phone:+81-29-861-9237  Fax:+81-29-861-3389  E-mail: n-tsurumachi@aist.go.jp

1. Introduction

Recently, much effort has been devoted to the coherent control of exciton population in semiconductor using phase-locked ultrashort laser pulse pairs [1]. Especially, excitons in semiconductor quantum dots (QDs) are suitable for the coherent control due to their very long phase relaxation time and large transition dipole moment compared with atomic systems [2], and they have been expected to be used as quantum bits of the quantum computing. For this purpose, single QD spectroscopy using a microscope objective system or a scanning near-field microscope system is required to avoid the large inhomogeneous broadening in the transition spectrum of QD ensembles. On the other hand, the QD ensemble will be necessary for other applications such as nonlinear optical devices in order to obtain large signal intensity. In this case, the usual coherent control of excitons using phase-locked double pulses is quite difficult because of the inhomogeneous broadening. In this paper, we propose a new coherent control method that is available even for inhomogeneously broadened self-assembled QD systems. In this method, incident light with a specific pulse area is used. We also describe its application for ultrafast optical devices.

2. Theory

We simulate the coherent control process of exciton population in a QD as a simple two-level system using the optical Bloch equation for homogeneously broadened system such as a single QD and for inhomogeneously broadened system such as a QD ensemble. The Bloch vector \(B\) represents a state of the two-level system. The population difference \(w\) and coherence \(\rho\) are proportional to \(B_x\) and \(B_x+iB_y\), respectively.

First, we consider the case of simple excitation by a phase-locked pulse pair whose relative phase is \(\pi\). The excited state population and the coherence do not exist before the excitation and the excited population increases and the coherence is created by the first pulse excitation. After the first pulse excitation, in the case of homogeneously broadened system, the quantity of the population and the coherence will remain for energy relaxation time \(T_1\) and phase relaxation time \(T_2\), respectively. The system can be de-excited compulsorily if the second pulse whose relative phase is \(\pi\), is incident during the coherence remains. The coherent control of exciton population in semiconductor quantum well or a single QD has been performed using this technique [1, 2].

In the case of the inhomogeneously broadened system, on the other hand, it is difficult to coherently control of excitons with such a simple phase-locked pulse pair since the macroscopic coherence will disappear before \(T_2\) due to rapid dephasing of the spectrally distributed macroscopic polarization. In order to overcome this problem, we propose new-type coherent control technique for inhomogeneously broadened systems using an area-regulated pulse train. Figure 1 shows one of the coherent control methods for inhomogeneously broadened system using an area-regulated pulse train.

(a) Initially, the system is in the ground state and all Bloch vectors of each polarization are \(B=\begin{pmatrix} 0 & 0 & -1 \end{pmatrix}\).

(b) At \(t=0\), the system is excited by pulse #1 whose pulse area is \(\pi/2\). The Bloch vectors rotate in y-z plane to \(B=(0 -1 0)\) if \(\Omega \gg \Delta\), where \(\Omega\) is the Rabi frequency and \(\Delta\) is the detuning between the incident light frequency \(\omega_{ex}\) and the exciton resonant frequency \(\omega_{ex}\).

(c) The macroscopic coherence disappears due to rapid dephasing of spectrally distributed macroscopic polarization. This fact gives rise to coherent emission in the form of free induction decay whose decay time \(T_2^*\) is inversely proportional to the inhomogeneous broadening spectral width \(\sigma\).

(d) At \(t=\tau\), the system is excited by pulse #2 whose pulse area is \(\pi\). All vectors rotate 180 degree around x-axis. The result is that all vectors undergo a mirror reflection about x-z plane.

(e) The macroscopic coherence is regenerated due to rephasing of macroscopic polarization at the same rate as that of the dephasing process. The value of all the vectors becomes equal to \(B=(0 1 0)\) at \(t=2\tau\). In the case of the 2-pulse photon echo, the signal is generated at this time.

(f) For coherent control of inhomogeneously broadened systems, the system is excited by pulse #3 whose pulse area is \(\pi/2\) at \(t=2\tau\). At this time, all the polarizations are in-phase, so that they can rotate together to \(B=(0 0 -1)\) and the system can be made back to the initial ground state compulsorily without restriction of lifetime. In this case, all the pulse should be in-phase.

Conditions required for this new-type coherent control
method using an area-regulated pulse train for inhomogeneously broadened systems are the following.

1. The sum of all the pulse areas should be a multiple of $2\pi$. (In Fig. 1 case, $\pi/2+\pi+\pi/2=2\pi$.)

2. The macroscopic dephasing process should be canceled by a rephasing process, for example, due to $\pi$ pulse excitation.

3. The phase relaxation time $T_2$ of each exciton polarization (not $T_2^*$) should be sufficiently long.

4. The Rabi frequency $\Omega$ should be considerably larger than the frequency detuning $\Delta$ between the incident light frequency $\omega_L$ and the exciton resonant frequency $\omega_{ex}$, or the inhomogeneous broadening width $\sigma$.

3. Numerical simulation

We performed numerical simulation of this process.

We assumed that the pulse width $\delta$ and the interval $\tau$ are 100 fs and 500 fs, respectively. The spectrum was assumed to have a Gaussian profile whose width is 10 or 100 meV. The phase relaxation time $T_2$ was assumed to be 100 ps. Figure 2 shows the time evolution of the coherence and the excited state population of the system obtained from the Bloch vector components. In the case of $\sigma=10$ meV, both the coherence and the population evolve in time almost exactly as the expected in the discussion in section 2. As a result, the system returns to the ground state almost completely after 1 ps. On the other hand, in the case of $\sigma=100$ meV, the Bloch vector behavior is different from the expectation and the system cannot return to the ground state. In this case, all the vectors do not rotate in the same way since the inhomogeneous broadening width $\sigma$ is so large that condition (4) of this method is not satisfied. However, one can coherently control the inhomogeneously broadened system with $\sigma=100$ meV using shorter laser pulses, for example $\delta=10$ fs.

This process is related with well-known Rabi-oscillation and self-induced transparency, and can be applied for ultrafast optical switches without restriction of the energy relaxation time $T_1$. By changing the pulse sequence, we can also expect four-wave mixing type ultrafast optical devices.

4. Conclusions

We have proposed a new coherent control method that is available even for inhomogeneously broadened systems, which uses an area-regulated laser pulse train. Its application for ultrafast optical devices without restriction of energy relaxation time is expected.

References