Coherent control of inhomogeneously broadened system by area-regulated pulse sequence

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(Received 1 August 2003; accepted 26 January 2004)

We have proposed a coherent control method that is available even for inhomogeneously broadened systems, which uses an area-regulated laser pulse sequence. It is expected to be applied to ultrafast optical devices without restriction of energy relaxation time. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1689743]

Ultrafast all-optical switching devices are very important for ultrafast optical communication systems and ultrafast information processing. For this purpose, it is necessary to find nonlinear optical materials with high optical nonlinearity and fast response. In general, however, there is a trade-off between the magnitude of nonlinear susceptibility and response time, and it is not easy to satisfy these two requirements at the same time.[1] Although semiconductor quantum nanostructures have been expected to be used in practical photonic devices because of their large optical nonlinearity, they are not suitable for ultrahigh repetition processing since their excited state lifetimes are generally on the order of several 100 ps.

In order to overcome this problem, recently much effort has been devoted to coherent control of the exciton population in semiconductors using phase-locked ultrashort laser pulse pairs.[2] As long as the coherence of the induced polarization by the first excitation pulse remains, the system can be returned to the initial ground state by the coherent control using the second excitation pulse whose relative phase is π, even if the nonlinear materials have long energy relaxation time.[3] Especially, excitons in semiconductor quantum dots (QDs) are suitable for coherent control due to their very long phase relaxation times and large transition dipole moments compared with those of atomic systems.[4,5] QDs have also been expected to be used as quantum bits in quantum computing.[6] 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.[7] On the other hand, QD ensembles will be necessary for applications in ultrafast nonlinear optical devices in order to obtain large signal modulation. In this case, the usual coherent control of excitons using phase-locked double pulses is quite difficult because of the inhomogeneous broadening.

In this letter, we propose a 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.

We simulate the coherent control process of exciton population in a QD as a simple two-level system using the optical Bloch equation[8] for a homogeneously broadened system such as a single QD and for an inhomogeneously broadened system such as a QD ensemble. The Bloch vector **B** represents the state of the two-level system. The population difference w and coherence ρ are proportional to B_z and $B_x + iB_y$, respectively.

First, we consider the case of simple excitation by a phase-locked pulse pair whose relative phase is π. The excited state population and the coherence are assumed not to exist before the excitation, and are created by the first pulse excitation. First, we consider homogeneously broadened systems. After the first pulse excitation, 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 forced into deexcitation if a second pulse with a relative phase π is applied while coherence remains. The coherent controls of the exciton population in semiconductor quantum wells and a single QD have been performed using this technique.[2,7,9]

With inhomogeneously broadened systems, on the other hand, it is difficult to coherently control excitons using such a simple phase-locked pulse pair since the macroscopic coherence will disappear in much shorter time than T_2. The rapid dephasing of the macroscopic polarization is determined by the inhomogeneous broadening of the spectral width. In order to overcome this problem, we propose a coherent control technique for inhomogeneously broadened systems using an area-regulated pulse sequence. Generally the area of a pulse is defined as $\int \mu E(t) dt/\hbar$, where $E(t)$ is electric field envelope and μ is the transition dipole moment. Pulses with an area of π and $\pi/2$ play an important role in photon echoes and related nonlinear coherent transient phenomena.[8] Figure 1 schematically shows one of the coherent control methods for inhomogeneously broadened systems using an area-regulated pulse sequence, which is described in the following.

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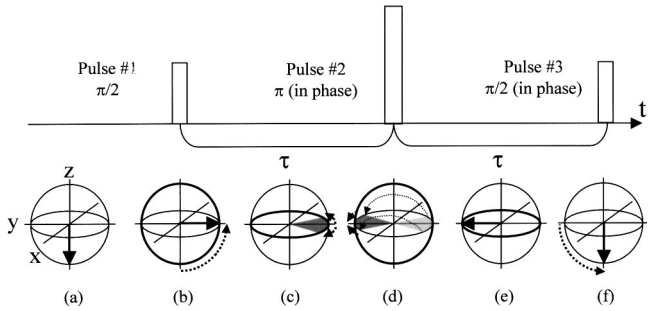

FIG. 1. Schematic drawings describing the coherent control procedure of inhomogeneously broadened systems. The upper picture shows the pulse excitation sequence. The lower picture depicts the precession of the Bloch vector at various times.

(a) Initially, the system is assumed to be in the ground state and all Bloch vectors of each two-level system are $\mathbf{B}=(0\ 0\ -1)$.
(b) At $t=0$, the system is excited by pulse 1 whose pulse area is $\pi/2$. The Bloch vectors rotate in the $y-z$ plane to $\mathbf{B}=(0\ -1\ 0)$ provided $\Omega \gg \Delta$, where $\Omega=\mu E(t)/\hbar$ is the Rabi frequency and Δ is the detuning between the incident light frequency ω_L and the exciton resonant frequency ω_{ex}.
(c) The macroscopic coherence disappears due to the rapid dephasing of the spectrally distributed macroscopic polarization. The macroscopic polarization gives rise to coherent emissions in the form of free induction decay whose decay time T_2^* is inversely proportional to the inhomogeneous broadening spectral width σ.
(d) At $t=\tau$, the system is excited by pulse 2 whose pulse area is π. All vectors rotate 180° around the x axis. The result is that all vectors undergo a mirror reflection about the $x-z$ plane.
(e) The macroscopic coherence is regenerated due to rephasing of the macroscopic polarization at the same rate as that of the dephasing process. The value of all the vectors becomes equal to $\mathbf{B}=(0\ 1\ 0)$ at $t=2\tau$ if the dephasing associated with homogeneous broadening is neglected. This is exactly the generation process of two-pulse photon echoes.
(f) For coherent control, or coherent damping, of the inhomogeneously broadened systems, the system is illuminated by pulse 3, whose pulse area is $\pi/2$, at $t=2\tau$. At the time just before pulse 3, all the polarizations are in-phase, so that they rotate together to $\mathbf{B}=(0\ 0\ -1)$ and the system is forced back to its initial ground state even if its lifetime is much longer. In the sequence above, all the pulses are set in-phase.

The conditions required for this coherent control method using an area-regulated pulse sequence for inhomogeneously broadened systems are as follows.

(1) The sum of all the pulse areas should be a multiple of 2π. (In Fig. 1 case, $\pi/2+\pi+\pi/2=2\pi$.)
(2) The macroscopic dephasing process should be reversed by a rephasing process, for example, by π pulse excitation.
(3) The phase relaxation time T_2 of each exciton polarization (not T_2^*) should be sufficiently long.

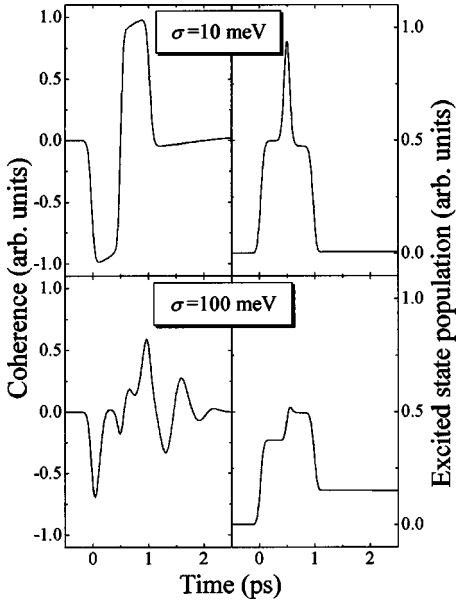

FIG. 2. The calculated time evolution of coherence (left) and the excited state population (right) of the system obtained from the Bloch vector components for the coherent control of inhomogeneously broadened systems in the case of $\sigma=10$ meV (upper) and 100 meV (lower). The excitation pulse sequence is shown in Fig. 1.

(4) The Rabi frequency Ω should be considerably larger than the inhomogeneous width σ.

We performed a numerical simulation of this process. We assumed that the pulse width δ and the interval τ are 100 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 expected in the above-presented discussion. As a result, the system returns to the ground state almost completely at 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 does not return to the ground state completely. In this case, all the vectors do not rotate in the same way since the inhomogeneous width σ is so large that condition (4) required for this method is not satisfied. However, one can coherently control the inhomogeneously broadened system having $\sigma=100$ meV using shorter laser pulses, for example $\delta=10$ fs. This process is related to well-known Rabi oscillations and self-induced transparency, and can be applied to ultrafast optical switches without restricting long energy relaxation time T_1.

We propose another pulse sequence, which can be applied to four-wave mixing-type ultrafast optical devices. Figure 3 shows the pulse sequence and an example of the simulation of this coherent control method for inhomogeneously broadened systems. We apply four pulses whose duration and bandwidth are 15 fs and 120 meV at $t=0$, 1 ps, 3 ps, and 4 ps and whose pulse areas are $\pi/2$, π, π, and $\pi/2$, respectively. The spectrum was assumed to have a Gaussian profile whose width is 30 meV. The phase relaxation time T_2 was assumed to be 100 ps. Note that the pulses 2 and 3 are in

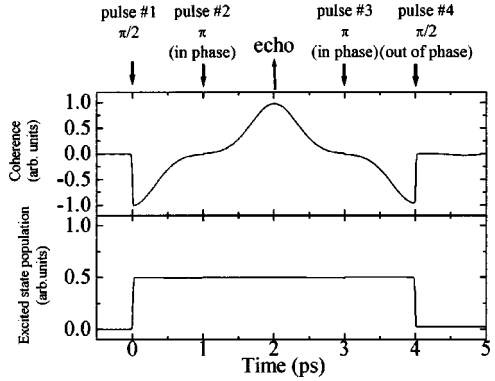

FIG. 3. Time evolution of coherence (upper) and the excited state population (lower) of the system obtained from the Bloch vector components for the photon echo-type coherent control in the case of $\sigma=30$ meV.

phase with pulse 1, and that pulse 4 is out of phase. In this case, we can expect the photon echo signal at $t=2$ ps. Finally, the excited state population of the system disappears almost completely and the system returns to the ground state. Although one can obtain the information of dephasing by changing all the pulse intervals, the dephasing dynamics may involve exciton–exciton scattering effect or other intensity dependent effects due to the comparatively large incident intensity. The main purpose of this method is to erase the residual excited state population after generating photon echo signal.

In this calculation, we assumed that the quantity of transition dipole moment μ is constant in the whole inhomogeneously broadened system. The dipole moment of each QD is distributed due to the size fluctuation in the case of self-assembled QDs. The pulse area depends on μ, therefore the ratio of the QDs which do not satisfy condition (1) increases. As a result, it is necessary to take this dipole moment distribution into account for the analysis of the performance in self-assembled QD system and the conditions of the coherent control using the area-regulated pulse sequence become more stringent. On the other hand, since the transition dipole moment is constant in the case of atomic systems, this method can be available if the dephasing time is sufficiently long.[10–12]

In summary, we have proposed coherent control methods that are available even for inhomogeneously broadened systems, which use an area-regulated laser pulse sequence. It is expected to be applied to ultrafast optical devices without restriction of energy relaxation time.

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