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Ultrafast Coherent Control of Inhomogeneously Broadened System by an Area-Regulated Pulse Sequence

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We have proposed a new 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. [DOI: 10.1143/JJAP.43.2023]

KEYWORDS: coherent control, inhomogeneously broadened system, self-assembled quantum dots, area-regulated pulse sequence

1. Introduction

Ultrafast nonlinear optical 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 simultaneously.[1)] Although semiconductor quantum nano structures have been expected to be used in practical photonic devices because of their large optical nonlinearity, they are not suitable for ultrahigh repetition processing in the THz region since their excited state lifetime is generally of the order of 100 ps or 1 ns.

In order to overcome this problem, considerable effort has recently been devoted to the coherent control of an exciton population in semiconductors using phase-locked ultrashort laser pulse pairs.[2)] In particular, excitons in semiconductor quantum dots (QDs) are suitable for coherent control due to their very long phase relaxation time and large transition dipole moment compared with those of atomic systems.[3,4)] QDs have also been expected to be used as quantum bits in quantum computing.[5)] For this purpose, single-QD spectroscopy using a microscope objective system or a scanning near-field microscope system is required to prevent large inhomogeneous broadening in the transition spectrum of QD ensembles.[6)] On the other hand, QD ensembles will be necessary for their applications in ultrafast nonlinear optical devices in order to obtain large signal modulation. In this case, the typical coherent control of excitons using phase-locked double pulses is quite difficult because of inhomogeneous broadening.

In this paper, we propose a new coherent control method that is available even for inhomogeneously broadened systems such as the QD ensemble. In this method, incident light with a specific pulse area is used. We also describe its application in ultrafast optical devices.

2. Coherent Control by a Phase-Locked Pulse Pair

First, coherent control by a phase-locked pulse pair is described. In the case of a homogeneously broadened system such as a single QD, this coherent control method is useful for ultrafast excited state carrier damping. 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 coherent control using the second excitation pulse whose relative phase is π, even if the energy relaxation time is long.[7)]

We simulate the coherent control of an 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 $\boldsymbol{B}$ represents the state of a two-level system. The population difference w and coherence ρ are proportional to B_z and $B_x + iB_y$, respectively.

The case of simple excitation by a phase-locked pulse pair, whose relative phase is π, is considered. We performed a numerical simulation of this process for a homogeneously broadened system and an inhomogeneously broadened system. We assumed that the pulse width δ and the interval τ are 100 fs and 1 ps, respectively. The spectrum of the inhomogeneously broadened system was assumed to have a Gaussian profile whose width is 30 meV. The phase relaxation time T_2 was assumed to be 100 ps. Figure 1 shows the time evolution of the coherence and the excited-state population of the system obtained from the Bloch vector components. The excited-state population and the coherence were assumed not to exist before the excitation, and the excited-state population and the coherence are created by the first pulse excitation. In the case of the homogeneously broadened system, after the first pulse excitation, the quantity of the population and the coherence remain almost constant for the energy relaxation time T_1 and the phase relaxation time T_2, respectively. The system can be forced into de-excitation if a second pulse, whose relative phase is π, is incident while the coherence remains. The coherent control of an exciton population in semiconductor quantum wells and a single QD has been performed using this technique.[2,6,9)]

With the inhomogeneously broadened system, on the other hand, the macroscopic coherence will disappear in a much shorter time than T_2. The rapid dephasing of the macroscopic polarization is determined by the inhomogeneous broadening of the spectrum. In this case, the coherent

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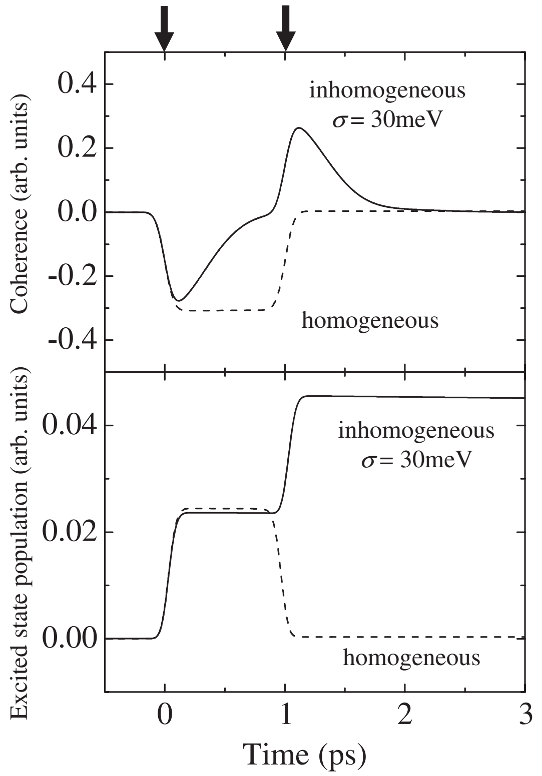

Fig. 1. Calculated time evolution of the coherence (top) and the excited state population (bottom) of the system obtained from Bloch vector components for the coherent control of homogeneously and inhomogeneously broadened systems in the case of $\sigma = 30$ meV by a phase-locked pulse pair. The timing of excitation pulses is denoted by arrows.

damping of excitons using such a simple phase-locked pulse pair is impossible, and the exciton population is increased by the second pulse excitation.

3. Coherent Control by an Area-Regulated Pulse Sequence

In order to overcome this problem, we propose a new coherent control technique for inhomogeneously broadened systems using an area-regulated pulse sequence. Generally, the area of a pulse is defined by $\int \mu E(t)\mathrm{d}t/\hbar$, where $E(t)$ is the electric field envelope and μ is the transition dipole moment. Pulses with areas of π and $\pi/2$ play an important role in photon echoes and related nonlinear coherent transient phenomena.[8)]

Figure 2 schematically shows one of the coherent control methods for inhomogeneously broadened systems using an area-regulated pulse sequence, which is described in the following.

(a) Initially, the systems are assumed to be in the ground state and the Bloch vector of each two-level system is $\boldsymbol{B} = (0, 0, -1)$.

(b) At $t = 0$, the systems are excited by pulse #1 whose area is $\pi/2$. The Bloch vectors rotate in the y-z plane to $\boldsymbol{B} = (0, -1, 0)$, providing $\Omega \gg \Delta$ 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 time $T_2{}^*$ is inversely proportional to the inhomogeneous broadening spectral width σ.

(d) At $t = \tau$, the systems are excited by pulse #2 whose area is π. All vectors rotate 180 degrees 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 the rephasing of the macroscopic polarization at the same rate as that of the dephasing process. The value of all vectors becomes $\boldsymbol{B} = (0, 1, 0)$ at $t = 2\tau$ when the dephasing associated with homogeneous broadening is neglected. This is exactly the generation process of two-pulse photon echoes.

(f) For the coherent control or coherent damping of the inhomogeneously broadened systems, the systems are illuminated by pulse #3, whose area is $\pi/2$, at $t = 2\tau$. At the time just before pulse #3 illumination, all polarizations are in-phase, so that they rotate together

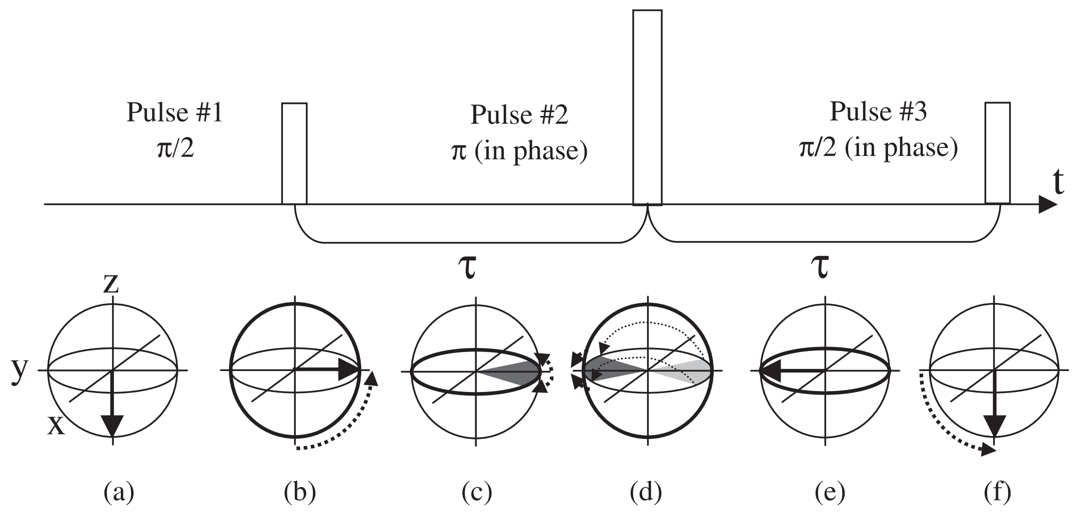

Fig. 2. Schematics describing the coherent control procedure of inhomogeneously broadened systems. The top diagram shows the pulse excitation sequence. The bottom diagram depicts the precession of the Bloch vector at various times.

to $\boldsymbol{B} = (0, 0, -1)$ and the systems are forced back to their initial ground state even if their lifetime is much longer than 2τ. In the sequence above, all pulses are set in-phase.

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

(1) The sum of all pulse areas should be a multiple of 2π. (In the case shown in Fig. 1, $\pi/2 + \pi + \pi/2 = 2\pi$.)
(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.
(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 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 3 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 that expected in the discussion above. 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 vectors do not rotate in the same manner 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, those with $\delta = 10$ fs, because the Rabi frequency of the shorter pulses is larger for the same pulse area. This process is related to well-known Rabi oscillations and self-induced transparency, and can be applied to ultrafast optical switches without restriction of long energy relaxation time T_1.

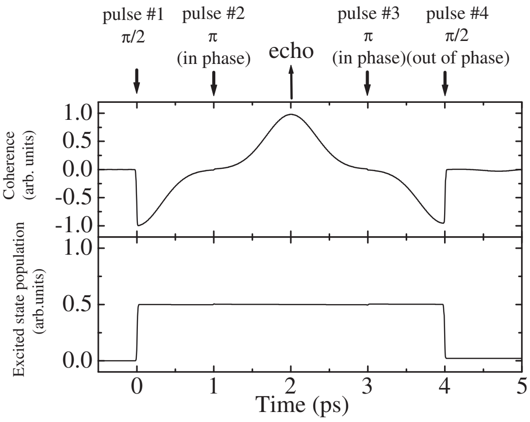

Fig. 4. Time evolution of coherence (top) and the excited state population (bottom) of the system obtained from Bloch vector components for photon echo-type coherent control in the case of $\sigma = 30$ meV.

We propose another pulse sequence, which can be applied to four-wave-mixing-type ultrafast optical devices. Figure 4 shows the pulse sequence and the result of the simulation of this coherent control method for inhomogeneously broadened systems. We apply four pulses whose duration is 15 fs at $t = 0$, 1 ps, 3 ps, and 4 ps and whose 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 pulses #2 and #3 are in-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.

The beam directions were assumed to be the same, namely, collinear. However, a noncollinear situation is also very important for image processing. Next, we discuss a noncollinear configuration. Figure 5 schematically shows one of the coherent control methods using an area-regulated pulse sequence in the case of a noncollinear configuration. Three pulses with different wave vectors, $\boldsymbol{k}_1$, $\boldsymbol{k}_2$, and $\boldsymbol{k}_3$, are simultaneously incident. In this case, we can expect to

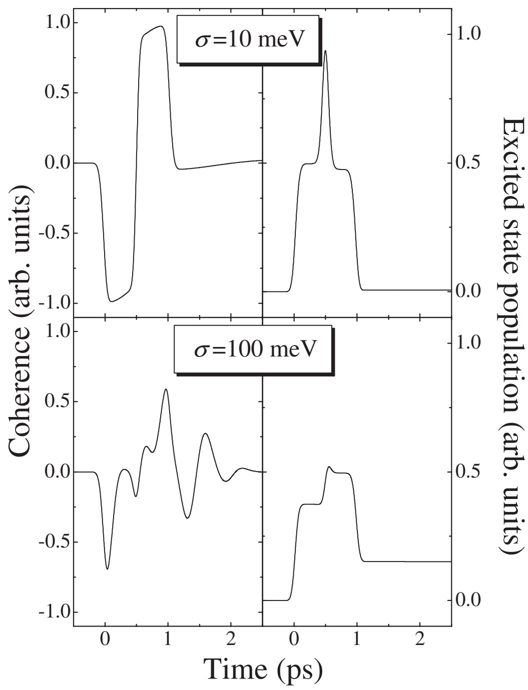

Fig. 3. Calculated time evolution of the coherence (left) and the excited state population (right) of the system obtained from Bloch vector components for the coherent control of inhomogeneously broadened systems in the case of $\sigma = 10$ meV (top) and 100 meV (bottom). The excitation pulse sequence is shown in Fig. 2.

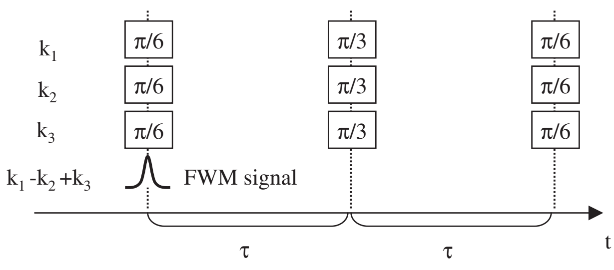

Fig. 5. Pulse sequence for coherent control methods using area-regulated pulses in the case of noncollinear configuration.

observe a four-wave-mixing signal with the wave vector $\boldsymbol{k}_1 - \boldsymbol{k}_2 + \boldsymbol{k}_3$ at this time. Our purpose is to rapidly erase the excited-state population compulsorily after this four-wave-mixing process. The areas of the first three pulses are $\pi/6$. In this case, the sum of the pulse area is $\pi/2$. Next, at $t=\tau$, three pulses are incident, whose wave vectors are $\boldsymbol{k}_1$, $\boldsymbol{k}_2$, and $\boldsymbol{k}_3$, and areas are $\pi/3$. Finally, $\pi/6$ area pulses are incident in these three directions. As a result, the system return to its initial ground state since the sum of all pulse areas is 2π. However, there is a problem in noncollinear excitation. Namely, when light beams with different directions overlap, an interference spatial fringe is generated in this overlap region. Therefore, the intensity of light is modulated periodically in this region. In this coherent control method, the pulse area of each position, which is quite an important parameter, is varied by this fringe. This leads to the imperfection of coherent control. In order to overcome this problem, we should use noncollinear beams with different polarizations. When the polarizations of two beams are cross-linear, there is no intensity modulation in the overlap region.[10] This differs from the situation where two beams have the same polarization. In this case, a polarization grating will be generated in this region and we can observe a four-wave-mixing signal. Therefore, when two of the three incident beams are counter propagated, which is well known as phase conjugation,[11] noncollinear coherent control is possible.

4. Conclusions

In summary, we have proposed a new 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.

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