Decoherence suppression of excitons in GaSe using three successive femtosecond optical pulses

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(Received 25 May 2006; published 30 August 2006)

We report the first decoherence control of excitons which was performed on the model semiconductor GaSe by using successive three femtosecond pulses, i.e., the six-wave mixing configuration. The second pulse acts as a π pulse which reverses the time evolution of non-Markovian dynamics. By changing the pulse interval conditions, we confirmed the suppression of exciton decoherence by π pulse irradiation. From the analysis of the decoherence, it is also found that the six-wave mixing technique can successfully be applied to determine the reservoir parameters.

DOI: 10.1103/PhysRevB.74.073202 PACS number(s): 78.47.+p, 03.67.Pp, 42.50.Md

Decoherence is one of the utmost important processes which gives information of the interaction between the electron system and the thermal reservoir. The decoherence process is widely investigated by the four-wave mixing (FWM) method, and is also studied by the six-wave mixing (SWM) technique which provides the information of many-body effects involving a higher order correlation.[1,2] Decoherence also attracts much attention for the technological advancement of quantum information. A central issue is how to control and suppress the decoherence of well coded quantum states. One practical method of decoherence control is to apply a sequence of state-reversing pulses (π pulses) on the system with their intervals being less than the noise correlation time. This is now referred to as "bang-bang" (BB) control.[3–6] The key idea is to reverse the time evolution of non-Markovian dynamics by using π pulses.[6,7] Due to this time reversibility in the non-Markovian regime, the contribution of various reservoir modes, such as thermally-induced phonon modes, is negated simultaneously, and the decoherence time can be extended up to the energy relaxation time T_1 by irradiating a sequence of π pulses. This method is relatively easy to implement compared to other methods such as quantum error correcting codes[8–10] and encoding with decoherence free subspace,[11] which require encoding with multiparticle entangled states. Therefore the BB control should be the first choice for testing decoherence control, and has been successfully applied to trapped atom systems[12] and solid-state nuclear spin systems.[13] However the BB control of charged states, such as excitons, in semiconductors has not been reported yet to our knowledge, although various results of optical coherent manipulation of excitons,[14,15] including the coherent control of the coupling of a particular LO phonon mode, have been reported.[16]

In this paper, we present the first experimental demonstration of the BB control of excitons in a semiconductor, suppressing the decoherence caused by the coupling with an unlimited number of reservoir modes. Excitons are clearly distinct from other two-level systems for qubits in the sense that they directly couple to photons, which are the signal carriers for building a communication network. We applied three femtosecond pulses onto excitons in layered semiconductor GaSe, and observed the decoherence suppression caused by the second, intermediate pulse. The technique developed in the present work can be extended to useful means not only for stabilizing charged qubits but also for identifying relevant decoherence characteristics in detail.

The sample in this study was a bulk ensemble of $1S$ excitons in layered semiconductor GaSe whose resonant energy is 2.11 eV at 5 K. All measurements were performed at 5 K. The excitons were driven resonantly by optical pulses generated by an optical parametric oscillator (OPO), with a repetition rate of 76 MHz and a pulse duration of ~120 fs, pumped synchronously by a mode-locked Ti:sapphire laser. The excitation power was ~4 $\mu J/cm^2$, for which the optical Stark shift of excitons might be negligible.[17] The original light pulse from the OPO was divided into three pulses, and sent into the sample from three different directions with wave-vectors $\mathbf{k}_1$, $\mathbf{k}_2$, and $\mathbf{k}_3$. Their pulse areas were θ_1, θ_2, and θ_3, respectively. The polarizations of three pulses were set clockwise circular, in order to eliminate the biexcitonic contribution to the multiwave mixing process. The notation used denotes a time ordered process: index 1, 2, and 3 indicates the first, second, and third pulse, respectively (Fig. 1). The time interval between pulses No. i and No. j is written as τ_{ij}. The first pulse ($\mathbf{k}_1$, θ_1) generates excitons in a superposition state $\cos\frac{\theta_1}{2}|0\rangle+\sin\frac{\theta_1}{2}|1\rangle$ at time $t_1=0$. The excitons soon couple to the reservoir, starting to decohere. The two succes-

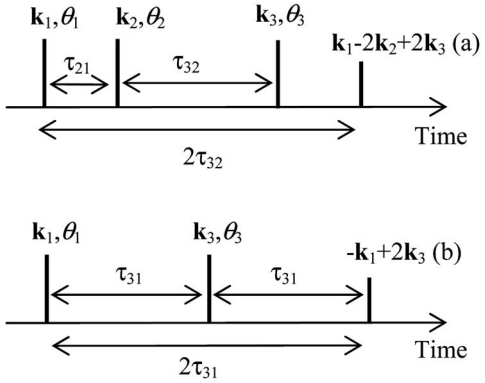

FIG. 1. The time evolutions of (a) SWM and (b) FWM. In the case of the SWM, the $\mathbf{k}_2$ pulse was added between the two pulses of the FWM. The SWM signal appears at $2\tau_{32}$, and the FWM signal appears at $2\tau_{31}$ from the first pulse.

sive pulses modify their quantum states, creating polarization lattices in the sample. These macroscopic polarizations of excitons radiate the multiwave mixing signals into certain directions. The decoherence can be measured by detecting these multiwave mixing signals, whose intensities directly reflect the intensities of the off-diagonal components of the density operator of exciton, i.e. $I(t) \propto |\rho_{10}(t)|^2$.

We are particularly interested in two multiwave mixing components; one is the SWM signal emitted in the direction $2\mathbf{k}_3 - 2\mathbf{k}_2 + \mathbf{k}_1$, and the other is the FWM signal emitted in the direction $2\mathbf{k}_3 - \mathbf{k}_1$. Their intensities are given by[18]

$$I_{2\mathbf{k}_3-2\mathbf{k}_2+\mathbf{k}_1}(t,\tau_{21},\tau_{32}) = \frac{1}{4}\sin^2\theta_1 \sin^4\frac{\theta_2}{2}\sin^4\frac{\theta_3}{2} \times \exp[-2\Gamma_{-+-}(t) - (t-2\tau_{32})^2\delta_B^2], \quad (1)$$

$$I_{2\mathbf{k}_3-\mathbf{k}_1}(t,\tau_{21},\tau_{32}) = \frac{1}{4}\sin^2\theta_1 \cos^4\frac{\theta_2}{2}\sin^4\frac{\theta_3}{2}\exp[-2\Gamma_{-++}(t) - (t-2\tau_{31})^2\delta_B^2], \quad (2)$$

$$\Gamma_{c_3c_2c_1}(t) = 2\int_0^\infty d\Omega I(\Omega)\frac{f_{c_3c_2c_1}(\Omega,t)}{\Omega^2}\coth\left(\frac{\hbar\Omega_1}{2k_BT}\right), \quad (3)$$

with

$$f_{c_3c_2c_1} = |c_3(e^{i\Omega t_3} - e^{i\Omega t}) + c_2(e^{i\Omega t_2} - e^{i\Omega t_3}) + c_1(e^{i\Omega t_1} - e^{i\Omega t_2})|^2, \quad (c_i = \pm 1), \quad (4)$$

and the inhomogeneous broadening of the exciton energies is taken into account assuming that it can be characterized by a Gaussian distribution with a variance δ_B. As can be seen from Eq. (1), the SWM echo signal appears at $2\tau_{32}$ away from the arriving time of the first $\mathbf{k}_1$ pulse [Fig. 1(a)].[19] This delay in the appearance of the echo was verified by the time-resolved SWM experiment, and found to be independent of the excitation density.[20] We also confirmed that the temporal profiles of the echo signals retain to a Gaussian shape up to our highest excitation densities. The exciton in the present system can thus be regarded as a two-level system.[21] According to the conventional phenomenological theory based on the relaxation time T_2, therefore, the exciton polarization created by the $\mathbf{k}_1$ pulse should decay as $\exp(-2\tau_{32}/T_2)$; thus the SWM signal intensity should decay monotonously as $\exp(-4\tau_{32}/T_2)$ with changing τ_{32}. In the case of the FWM signal, the arriving time of the second $\mathbf{k}_2$ pulse should have no influence on the process and the echo signal appears at $2\tau_{31}$. Within the framework of the T_2 theory, the FWM signal shows the well-known dependence of $\exp(-4\tau_{31}/T_2)$. In the present experiment, it is possible to select between the SWM and FWM processes simply by choosing the observation direction without changing any other conditions. Therefore, the pump intensity and the measurement temperature (5 K) are the same in both experiments.

In the ideal π-pulse case, $\theta_2 = \theta_3 = \pi$ only the SWM signal given by Eq. (1) must remain. The FWM signal given by Eq. (2), for example, vanishes. In our experiment, however, the pulse area is smaller than π, and is estimated to be $\theta_1 = \theta_2 = \theta_3 \sim \pi/2$ from the two pulse photon echo experiment. Thus, other multiwave mixing signals also appear, and the intensity of the SWM signal is reduced by a factor of $\frac{1}{4}\sin^2\theta_1 \sin^4\frac{\theta_2}{2}\sin^4\frac{\theta_3}{2}$ as shown in Eq. (1). Here we note that the decoherence exponent $\Gamma_{-+-}(t)$ in Eq. (1) does not change by what areas the irradiated pulses have. Therefore, we should be able to observe the decoherence suppression effect due to the ideal π pulses, which is included in $\Gamma_{-+-}(t)$, even if the irradiated pulses have an area smaller than π. In other words, the fraction $\frac{1}{4}\sin^2\theta_1 \sin^4\frac{\theta_2}{2}\sin^4\frac{\theta_3}{2}$ of excitons can effectively feel the ideal π-pulses, and this fraction of excitons can be time-reversed. On the other hand, the FWM signal given by Eq. (2) gives the information of decoherence in the two pulse case $(\theta_1, \theta_2, \theta_3) = (\pi/2, 0, \pi)$, which corresponds to the conventional photon echo scheme. The decoherence exponent $\Gamma_{-++}(t)$ is not affected by the second $\mathbf{k}_2$ pulse, and hence does not include the time-reversal process by this second pulse.

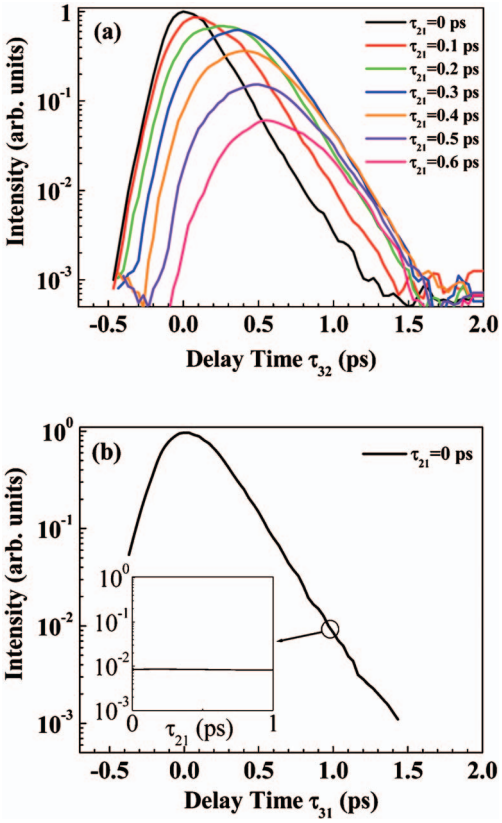

FIG. 2. (Color) (a) The τ_{21} dependence of the τ_{32} scanned SWM profile. The decay profile shifts toward longer delays of τ_{32} as the interval τ_{21} gets longer. (b) The FWM signal taken in the $-\mathbf{k_1} + 2\mathbf{k_3}$ direction. The inset shows that the signal intensity at $\tau_{31} = 1$ ps is not affected by the second pulse.

Figure 2(a) shows the signal profiles of the SWM as a function of τ_{32} obtained for several values of τ_{21}. According to conventional phenomenological theory based on the dephasing time T_2, the signal profiles should decay monotonously as $\exp(-4\tau_{32}/T_2)$, and should be independent of τ_{21}, i.e., of the timing of second $\mathbf{k}_2$ pulse. In the experiment,

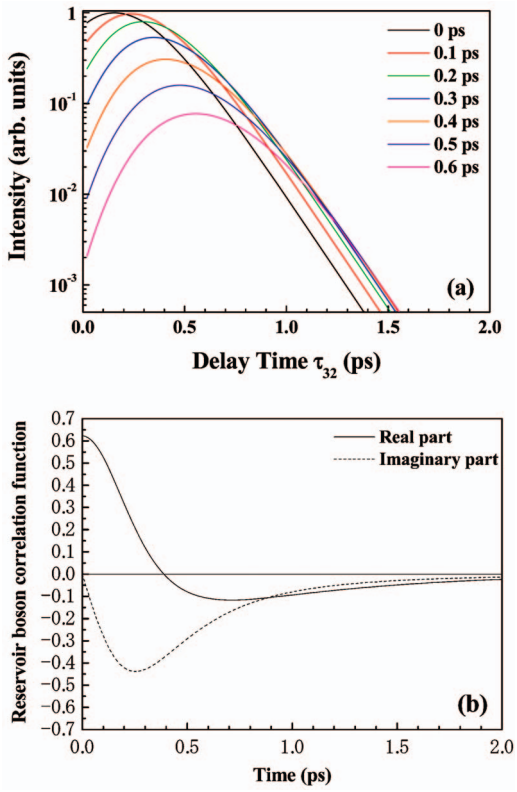

FIG. 3. (Color) (a) The calculation of SWM intensity. This represents the experimental result. The suppression of decoherence is confirmed. (b) Real and imaginary parts of the reservoir boson correlation function.

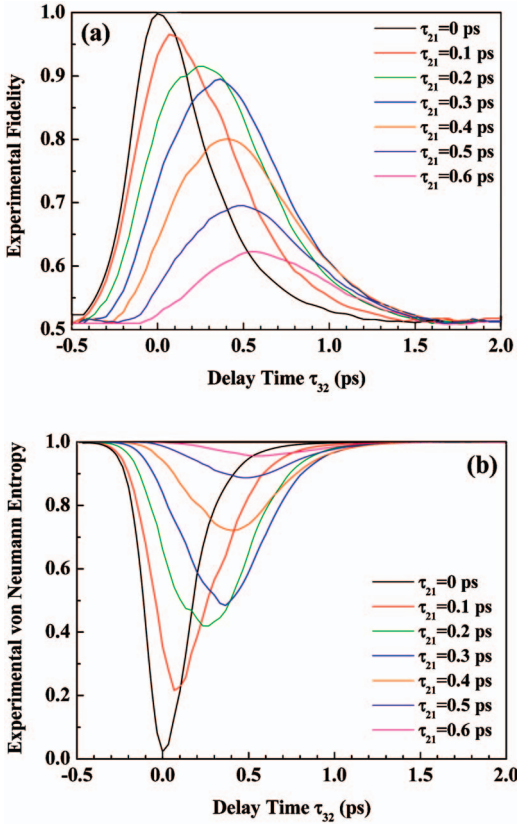

FIG. 4. (Color) The decay of purity of the initial quantum state of the exciton in terms of (a) the fidelity and (b) the von Neumann entropy.

however, the decay profile shifts toward longer delays of τ_{32} as the interval τ_{21} becomes longer. Namely, the decay is suppressed as the incident timing of the $\mathbf{k}_2$ pulse is delayed. This directly represents the decoherence suppression due to the time reversal by the $\mathbf{k}_2$ pulse in the non-Markovian regime. Ideally, the decoherence should not occur during the period of $2\tau_{21}$. The effect of time reversal is, however, imperfect because the time region is approaching the crossover from the non-Markovian to the Markovian regime. The shift of the decay profiles actually saturates for $\tau_{21} > 0.4$ ps. On the other hand, the decay profile of the FWM echo obtained under the three-pulse irradiation is unaffected by the temporal position of the $\mathbf{k}_2$ pulse, as expected by the expression of the decoherence exponent $\Gamma_{-++}(t)$ in Eq. (2). Figure 2(b) shows the decay profile of the FWM signal as a function of τ_{31}. In contrast to the SWM signals, the decay profile does not shift toward longer delays as the interval τ_{21} becomes longer.

Such behavior can be well reproduced by the fully quantized model of decoherence described by an exciton (qubit)-reservoir interaction Hamiltonian[18,22]

$$\hat{H}_{\mathrm{QR}} = \hbar \hat{\sigma}_z \sum_l (g_l \hat{B}_l^+ + g_l^* \hat{B}_l), \tag{5}$$

where $\hat{\sigma}_z$ is the z component of the Pauli matrix for the qubit ($\hat{\sigma}_z|0\rangle = -|0\rangle, \hat{\sigma}_z|1\rangle = |1\rangle$), $\hat{B}_l^+$ and $\hat{B}_l$ are the creation and annihilation operators of the reservoir boson in the lth mode, and g_l is the coupling constant for which the spectral density is defined by $I(\Omega) = \Sigma_l \delta(\Omega - \Omega_l)|g_l|^2$. We have adopted the ohmic spectral intensity for the reservoir boson modes

$$I(\Omega) = \alpha \Omega \exp\left(-\frac{\Omega}{\Omega_c}\right). \tag{6}$$

The best fitting parameters are extracted as $\Omega_c = 1.5$ meV, $\alpha = 0.3$, $\delta_B = 2$ meV, at $T = 5$ K. By using these parameters, we calculated the integrated SWM intensity $I^{\mathrm{int}}_{2\mathbf{k}_3 - 2\mathbf{k}_2 + \mathbf{k}_1}(\tau_{21}, \tau_{32}) = \int_{-\infty}^{\infty} dt I_{2\mathbf{k}_3 - 2\mathbf{k}_2 + \mathbf{k}_1}(t, \tau_{21}, \tau_{32})$ as a function of τ_{32} for several values of τ_{21}. The calculated SWM signal profile is shown in Fig. 3(a). The calculation reproduces perfectly the observed dependence of the SWM signal. We also evaluate the correlation function of the reservoir bosons $\langle \hat{X}(t)\hat{X}(0)\rangle$, where $\hat{X}(t) = \Sigma_l (g_l \hat{B}_l^+ e^{i\Omega_l t} + g_l^* \hat{B}_l e^{-i\Omega_l t})$ as shown in Fig. 3(b). The imaginary part peaks around $\tau_c \sim 0.4$ ps, implying that the boson modes suffer a large damping, and their correlation decays in this time region. This time scale coincides with the time scale at which the shift of the decay profile of the SWM signals saturates.

Finally, we analyze the rate of the purity degradation of the excitonic quantum state. Assuming an initial state $|\rho(0)\rangle = \frac{1}{\sqrt{2}}(|0\rangle + |1\rangle)$ created by the $\theta_1 \sim \pi/2$ pulse, the fidelity between the initial and the decohered exciton state is given by $F(t) = \langle \rho(0)|\hat{\rho}(t)|\rho(0)\rangle = \frac{1}{2}\{1 + \exp[-\Gamma_{-+-}(t)]\}$. The decoherence term $\exp[-\Gamma_{-+-}(t)]$ at $t = 2\tau_{32}$, i.e., the

peak position of the photon echo, can be evaluated experimentally by using the measured echo intensity, i.e., $\sqrt{I^{\mathrm{int}}_{2\mathbf{k}_3-2\mathbf{k}_2+\mathbf{k}_1}(\tau_{21},\tau_{32})}/\max I^{\mathrm{int}}_{2\mathbf{k}_3-2\mathbf{k}_2+\mathbf{k}_1}(\tau_{21},\tau_{32})$. Figure 4 shows the experimental fidelity thus obtained. We also plot the purity degradation measured by the von Neumann entropy $E=-F(2\tau_{32},\tau_{21},\tau_{32})\log_2 F(2\tau_{32},\tau_{21},\tau_{32}) -[1-F(2\tau_{32},\tau_{21},\tau_{32})]\log_2[1-F(2\tau_{32},\tau_{21},\tau_{32})]$, which must be zero for the pure state and unity for the completely decohered state. It can clearly be seen that the purity degradation is suppressed by applying the second $\mathbf{k}_2$ pulse within the reservoir correlation time.

Our results clearly demonstrate that the decoherence of excitons in a semiconductor can be suppressed by the BB control. The SWM scheme can provide a direct measurement of the reservoir correlation time τ_c. The method, however, needs further improvements. One immediate extension should be to increase the number of pulses. In the SWM scheme using three pulses, one can use the time-reversal of exciton states only once; then the exciton decoherence time can be extended up to the reservoir correlation time τ_c. If one applies pulses more frequently in a short time interval, the decoherence can be suppressed even longer.[3–6] Our method based on the multiwave mixing spectroscopy will be a powerful tool to investigate the so called non-Markovian dynamics which is less understood but essential for realizing coherent optical manipulation of exciton systems. The results obtained using this method will give more precise information on such reservoir characteristics.

We would like to thank C. Uchiyama and M. Ban for valuable discussions. This work was supported by a 21st Century COE Program at Tokyo Tech, by the Grant-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science, and Technology of Japan, and by the Strategic Information and Communications R&D Promotion Scheme.

[1] A. Lohner, K. Rick, P. Leisching, A. Leitenstorfer, T. Elsaesser, T. Kuhn, F. Rossi, and W. Stolz, Phys. Rev. Lett. **71**, 77 (1993).
[2] S. R. Bolton, U. Neukirch, L. J. Sham, D. S. Chemla, and V. M. Axt, Phys. Rev. Lett. **85**, 2002 (2000).
[3] M. Ban, J. Mod. Opt. **45**, 2315 (1998).
[4] L. Viola and S. Lloyd, Phys. Rev. A **58**, 2733 (1998).
[5] L. M. Duan and C. G. Guo, Phys. Lett. A **261**, 139 (1999).
[6] C. Uchiyama and M. Aihara, Phys. Rev. A **66**, 032313 (2002).
[7] M. Aihara, Phys. Rev. B **25**, 53 (1982).
[8] P. W. Shor, Phys. Rev. A **52**, R2493 (1995).
[9] A. M. Steane, Phys. Rev. Lett. **77**, 793 (1996).
[10] A. R. Calderbank and P. W. Shor, Phys. Rev. A **54**, 1098 (1996).
[11] D. A. Lidar, I. L. Chuang, and K. B. Whaley, Phys. Rev. Lett. **81**, 2594 (1998).
[12] M. F. Andersen, A. Kaplan, T. Grünzweig, and N. Davidson, Phys. Rev. A **70**, 013405 (2004).
[13] T. D. Ladd, D. Maryenko, Y. Yamamoto, E. Abe, and K. M. Itoh, Phys. Rev. B **71**, 014401 (2005).
[14] A. Schulzgen, R. Binder, M. E. Donovan, M. Lindberg, K. Wundke, H. M. Gibbs, G. Khitrova, and N. Peyghambarian, Phys. Rev. Lett. **82**, 2346 (1999).
[15] N. H. Bonadeo, J. Erland, D. Gammon, D. Park, D. S. Katzer, and D. G. Steel, Science **282**, 1473 (1998).
[16] M. U. Wehner, M. H. Ulm, D. S. Chemla, and M. Wegener, Phys. Rev. Lett. **80**, 1992 (1998).
[17] C. Hirlimann, J. F. Morhange, M. A. Kanehisa, A. Chevy, and C. H. Brito Cruz, Appl. Phys. Lett. **55**, 2307 (1989).
[18] M. Sasaki, A. Hasegawa, J. Ishi-Hayase, Y. Mitsumori, and F. Minami, Phys. Rev. B **71**, 165314 (2005).
[19] A. Hasegawa, T. Kishimoto, Y. Mitsumori, M. Sasaki, and F. Minami, J. Lumin. **108**, 211 (2004).
[20] T. Kishimoto, A. Hasegawa, Y. Mitsumori, J. Ishi-Hayase, M. Sasaki, and F. Minami, J. Lumin. (to be published).
[21] F. Jahnke, M. Koch, T. Meier, J. Feldmann, W. Schäfer, P. Thomas, S. W. Koch, E. O. Göbel, and H. Nickel, Phys. Rev. B **50**, 8114 (1994).
[22] G. M. Palmat, K. -A. Suominen, and A. K. Ekert, Proc. R. Soc. London **452**, 567 (1996).