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Decoherence suppression of excitons by bang–bang control

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Abstract

We report the demonstration of decoherence control of excitons on a layered compound 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 for the first time the suppression of exciton decoherence by π pulse irradiation.

Keywords: Decoherence; Six-wave mixing; Four-wave mixing; Photon echo

Decoherence is one of the most important processes which gives information on the interaction between the electron system and the thermal reservoir. It also attracts more attention for the technological advancement of quantum information. A central issue is how to control and suppress the decoherence of well-coded quantum states. The practical method might be the active dephasing control by using external pulsed-field sequences [1–3]. This is referred to as "bang–bang" (BB) control. The key idea is to reverse the time evolution of non-Markovian dynamics by optical π pulses [3,4]. Here, we present the experimental demonstration of the BB control of excitons in a semiconductor. The experiment was performed on excitons in a layered-semiconductor GaSe by a six-wave mixing (SWM) technique which consists of successive three pulses. The decoherence suppression caused by the second pulse was observed.

The excitation source was an optical parametric oscillator, with pulse duration of ~120 fs, pumped synchronously by a mode-locked Ti:sapphire laser. The center of the excitation energy is resonant with the 1S exciton (2.11 eV) in GaSe. All measurements were performed at 5 K. The original light was divided into three beams, and irradiated on the sample from three different directions with wave vectors $\boldsymbol{k}_1$, $\boldsymbol{k}_2$, and $\boldsymbol{k}_3$. The time interval between pulses $\sharp i$ and $\sharp j$ is written as τ_{ij} (Fig. 1). When the first pulse generates excitons, the excitons soon couple to the reservoir, starting to decohere. The two successive pulses modify the exciton quantum states. The modified polarizations of the excitons radiate the multi-wave mixing signals. The decoherence process can be measured by detecting these signals.

We are particularly interested in two multi-wave mixing signal components, the SWM signal emitted in the $\boldsymbol{k}_1-2\boldsymbol{k}_2+2\boldsymbol{k}_3$ direction and the four-wave mixing (FWM) signal emitted in the $-\boldsymbol{k}_1+2\boldsymbol{k}_3$ direction. In the experiment, it is possible to select the SWM and FWM processes simply by choosing the detected direction without changing any other conditions. In the case of SWM, the second pulse works as a π pulse like the second pulse of FWM. In the inhomogeneous broadening case, the SWM echo signal appears $2\tau_{23}$ away from the arriving time of $\sharp 1$ pulse under the condition of $\tau_{23}>\tau_{12}$ [5]. Therefore, the decay profile of coherence is observed by changing τ_{23}. We confirmed the signal generation condition (Fig. 2). In the conventional phenomenological theory based on the phase relaxation time T_2, the SWM signal intensity should decay monotonically as $\exp(-4\tau_{23}/T_2)$ with changing τ_{23}. In the case of FWM, the signal intensity shows the well-known dependence of $\exp(-4\tau_{13}/T_2)$ as a function of τ_{13}.

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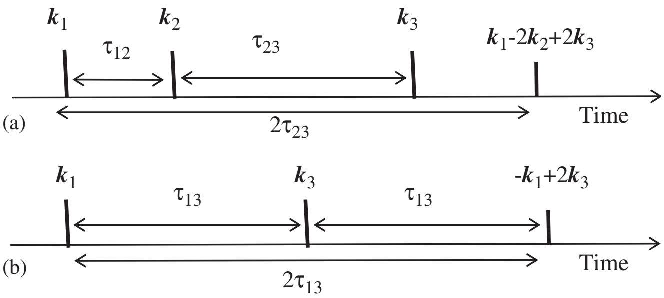

Fig. 1. The time evolutions of (a) SWM and (b) FWM. In the SWM configuration, the $\boldsymbol{k}_2$ pulse is added between the two pulses of the FWM. The SWM signal appears at $2\tau_{23}$, and the FWM signal appears at $2\tau_{13}$ from ♯1 pulse.

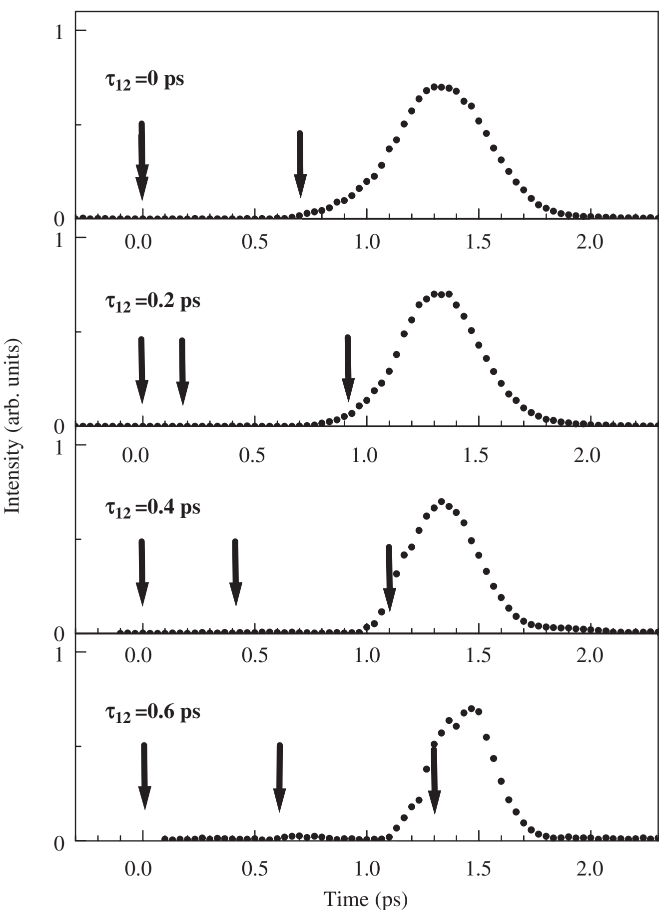

Fig. 2. The time-resolved SWM signals. τ_{23} is fixed at 0.7 ps. Time origin is the incident time of ♯1 pulse. Arrows indicate the incident time of pulses. The echo signals appear at 1.4 ps. This result shows that SWM signal is generated at $2\tau_{23}$.

For the ideal π pulse irradiation, only the SWM signal in the $\boldsymbol{k}_1 - 2\boldsymbol{k}_2 + 2\boldsymbol{k}_3$ direction remains and the FWM signal, e.g., vanishes. Since in our experiment, however, the pulse areas are smaller than π, other multi-wave mixing signals also appear, and the intensity of the SWM signal is reduced. Here, note that the decoherence process is not influenced by the areas of the pulses [6].

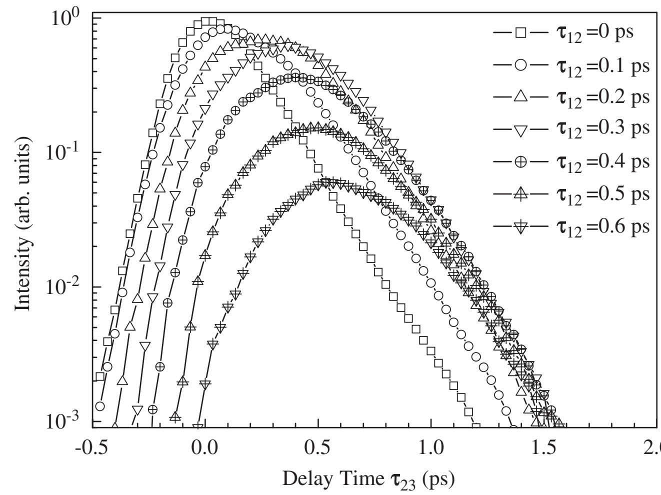

Fig. 3. The τ_{12} dependence of the τ_{23} scanned-SWM profile. The decay profile shifts toward longer delays of τ_{23} as τ_{12} gets longer. The delay time that it takes for the signals to decay from the initial intensity ($\tau_{12} = \tau_{23} = 0$ ps) to the same intensity lengthens, upon increasing τ_{12}.

Fig. 3 shows the signal profiles of SWM as a function of τ_{23} for several values of τ_{12}. According to the phenomenological theory, the signal profiles should decay monotonically, independent of τ_{12}. So, the experimental curves should overlap. In the result, however, the decay profile shifts toward longer delays of τ_{23} as τ_{12} becomes longer. Namely, the decay is suppressed during the interval τ_{12}. This directly represents the decoherence suppression due to the time reversal by ♯2 pulse in the non-Markovian regime. The effect of time reversibility, however, does not act when the time region is over the non-Markovian regime. The shift of the decay profiles actually saturates for $\tau_{12} > 0.4$ ps. On the other hand, we have confirmed that the decay profile of the FWM signal under the three pulse irradiation is unaffected by the temporal position of ♯2 pulse.

The decoherence of excitons arises from the random motion of the reservoir, which rapidly modulates the exciton transition frequency and causes degradation of the phase coherence. To simulate the result, we made a simple calculation based on the stochastic model. In the stochastic model, taking into account the finiteness of the reservoir correlation time τ_c, the SWM signal intensity is written as

$$I_{\mathrm{SWM}}(t, \tau_{12}, \tau_{23}) \propto \exp\left[-\{t - 2\tau_{23}\}\gamma_i^2\right]\left|\left\langle \exp\phi(t, \tau_{12}, \tau_{23})\right\rangle\right|^2,$$

$$\phi(t, \tau_{12}, \tau_{23}) = i\int_0^{\tau_{12}} \delta\omega(t)\,\mathrm{d}t - i\int_{\tau_{12}}^{\tau_{23}} \delta\omega(t)\,\mathrm{d}t + i\int_{\tau_{23}}^{t} \delta\omega(t)\,\mathrm{d}t,$$

where γ_i is the inhomogeneous broadening width and $\delta\omega(t)$ is the frequency modulation caused by the exciton–reservoir interaction. We assume the correlation function as

$$\left\langle \delta\omega(t_1)\delta\omega(t_2)\right\rangle = \Delta^2 \exp\left[-|t_2 - t_1|/\tau_c\right],$$

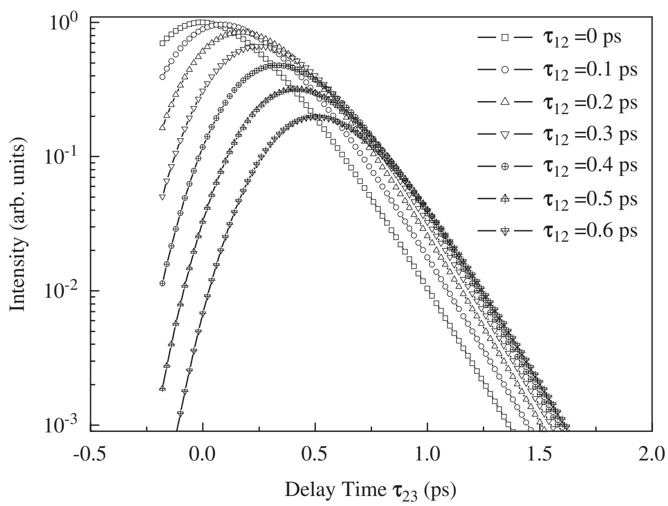

Fig. 4. The calculation of the SWM signal intensity. The parameters are $\tau_c = 0.3$ ps and $\Delta = 2.4$ THz.

where Δ is the amplitude of the frequency modulation. By taking the experimental value of γ_i and adjusting the parameters τ_c and Δ, we calculated the integrated SWM intensity as a function of τ_{23} for several values of τ_{12} (Fig. 4). The calculation reproduces perfectly the observed dependence of the SWM signal, which could be also confirmed by a more microscopic model [6]. The modulation by the reservoir cannot be regarded as a random process when the observation time scale is much shorter than the reservoir correlation time τ_c. Then the frequency modulation due to the reservoir is almost static and plays the role similar to that of the inhomogeneous broadening [4]. Therefore, ♯2 pulse causes some of the excitons to precess back towards their initial states, even if the excitons experience a frequency shift due to the motion of the reservoir.

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. In the SWM scheme using three pulses the exciton coherence time can be extended by the reservoir correlation time. If one applies pulses more frequently, the decoherence can be suppressed even longer. The multi-wave mixing spectroscopy will be a powerful tool to investigate the so-called non-Markovian dynamics which is less understood. The results obtained using this method will give more precise information on such reservoir characteristics.

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