Long-Time Correlation in Non-Markovian Dephasing of an Exciton-Phonon System in InAs Quantum Dots

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We have observed a time-correlated frequency fluctuation in non-Markovian dephasing of excitons in InAs quantum dots using a six-wave mixing technique. In this measurement, the arrival times of the excitation pulses were controlled to eliminate the influence of Markovian dephasing and to measure the pure non-Markovian behavior. The experimental result shows that the time correlation of the frequency fluctuation due to exciton-phonon interactions was maintained in the quantum dots for over 10 ps. This long-time correlation is caused by the modification of the phonon coupling distribution.

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Electrons in condensed matter are strongly influenced by various interaction processes, e.g., many-body and electron-phonon interactions. These interactions are investigated precisely by measuring the coherent dynamics of optically excited electrons. Four-wave mixing (FWM) measurements are commonly used to measure the coherent dynamics. In semiconductors, excitonic dephasing has been investigated in terms of exciton-exciton and exciton-phonon interactions [1–5]. The decrease in the exponential decay constant T_2 is well explained by a change in the instantaneous scattering rate based on the Markovian approximation. This dynamics is called Markovian dephasing. However, the initial dephasing process, before reaching Markovian dephasing, cannot be explained by phenomenological exponential decay. Time-correlated scatterings cause a more complicated decay, i.e., non-Markovian dephasing [6,7].

The transition from non-Markovian to Markovian dephasing is characterized by the loss of time correlation in the frequency fluctuation of the resonance energy. Since this transition is a fundamental aspect of microscopic dephasing, the non-Markovian behavior can be measured for a wide range of materials including gases, liquids, and solids [8–19]. Through the recent development of two-dimensional correlation spectroscopy, many-body correlations in semiconductors are being actively investigated [20,21]. Since the two-dimensional spectrum is sensitive to the phase modulation, it enables analysis of the detailed process of many-body correlations. Furthermore, the non-Markovian process attracts attention from the viewpoint of decoherence suppression, i.e., a reversing technique of the time evolution of the dephasing process by a sequence of optical pulses [22–24]. Control of the decoherence process has been achieved in some materials [25–27]. As shown in those investigations, to understand and control the interaction process, it is essential that the detailed mechanism of the coherent dynamics is resolved.

Since excitons in semiconductor quantum dots (QDs) exhibit long coherence time, i.e., nanosecond-scale Markovian dephasing, QDs are expected to show long-time non-Markovian dephasing. The extension of time correlation in non-Markovian dephasing makes it possible to reveal the fundamental interaction mechanism and leads to control of a multidegree quantum system. However, long-time non-Markovian dephasing has not yet been observed. It is difficult to observe the pure non-Markovian processes using conventional FWM measurements, since the inhomogeneity of the Markovian dephasing covers the non-Markovian nature. The Markovian dephasing for each QD varies widely due to the differences in confinement potential and coupled environment. To understand the microscopic interaction processes in QDs and other quantum-correlated systems, it is necessary to develop a measurement technique that is independent of the influence of Markovian dephasing.

In this Letter, we demonstrate the time-correlated frequency fluctuation of excitons in InAs QDs. To measure the pure non-Markovian dynamics, we used a six-wave mixing (SWM) technique. In the SWM measurement, time correlation could be observed clearly by eliminating the intensity change due to Markovian dephasing.

The investigated sample was grown on a InP(311)B substrate by molecular beam epitaxy. The sample contains 60 layers of InAs self-assembled QDs separated by 20-nm-thick $In_{1-x-y}Ga_xAl_yAs$ spacers. To fabricate highly stacked InAs QDs, the composition of the spacer layers was adjusted to compensate the strain in the InAs QD layers [28–30]. The FWM and SWM measurements were performed using optical pulses, generated by an optical parametric oscillator pumped by a mode-locked Ti:sapphire laser. The pulses were tuned to the exciton resonance wavelength of 1470 nm,

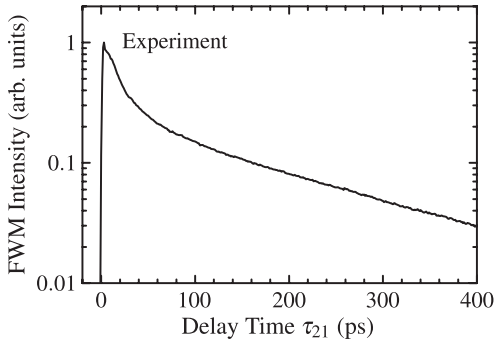

FIG. 1. Experimental four-wave mixing intensity for delay times of 0 to 400 ps.

the pulse duration was 130 fs, and the repetition rate was 76 MHz. The sample was mounted in a closed-cycle refrigerator. The first, second, and third excitation pulses were incident on the sample with wave vectors $\boldsymbol{k}_1, \boldsymbol{k}_2$, and $\boldsymbol{k}_3$, respectively. The FWM (SWM) signal in the direction of $2\boldsymbol{k}_2 - \boldsymbol{k}_1$ ($2\boldsymbol{k}_3 - 2\boldsymbol{k}_2 + \boldsymbol{k}_1$) was detected by a photodetector in transmission geometry with two-pulse (three-pulse) excitation. The signals were measured by changing the time interval between the first and second (second and third) excitation pulses, which is denoted by the delay time τ_{21} (τ_{32}). The excitation pulses were set to linear polarization along the [$\bar{2}33$] direction [30].

The FWM intensity at 3.4 K is shown in Fig. 1. The dephasing shape over 100 ps exhibits an exponential decay. It is well explained by a phenomenological T_2 decay based on the Markovian approximation. The initial dephasing within 100 ps shows a more rapid decay than the exponential T_2 decay. This rapid decay is expected to be caused by a time-correlated frequency fluctuation in the non-Markovian dephasing. The loss of the time correlation causes a gradual change from nonexponential to exponential decay. However, it is difficult to conclude that the nonexponential decay directly corresponds to the frequency fluctuation in the non-Markovian dephasing, since the FWM measurement cannot eliminate the influence of the ensemble of different Markovian T_2 decays. The Markovian dephasing, including the inhomogeneity of T_2, can show a nonexponential decay similar to the experimental result, e.g., double exponential decay, even though the time correlation is lost in the Markovian dephasing. The time correlation in non-Markovian dephasing is not sufficiently analyzed by the FWM signals. In order to reveal whether the initial decay is caused by the frequency fluctuation or not, it is necessary to do a measurement that is free from the inhomogeneity of T_2.

To measure the time correlation in the non-Markovian dephasing, we performed SWM measurements in which the intensity change due to the Markovian dephasing is eliminated. The SWM measurement is closely related to the decoherence suppression technique [27]. The illustration of this measurement is shown in Fig. 2(a). After the

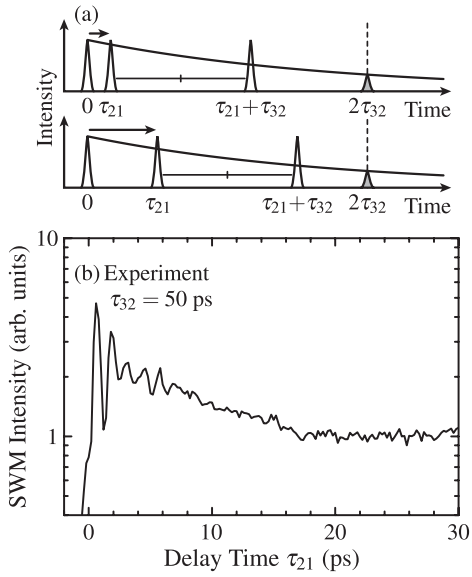

FIG. 2. (a) Illustration of the six-wave mixing measurement. The six-wave mixing intensity is measured as a function of the delay time τ_{21} with a fixed delay time τ_{32}. The upper and lower figures show the short and long conditions of the delay time τ_{21}, respectively. Photon echo signals appear at $2\tau_{32}$ for both conditions. (b) Experimental six-wave mixing intensity for the first delay times of $\tau_{21} = 0$ to 30 ps. The second delay time τ_{32} was set to 50 ps.

three-pulse excitation, the photon echo signal appears at $2\tau_{32}$. The SWM intensity in the Markovian dephasing shows an exponential decay $e^{-(4/T_2)\tau_{32}}$ corresponding to the rephasing time $2\tau_{32}$. Since the rephasing time depends on the second delay time τ_{32} only, the Markovian intensity is independent of the first delay time τ_{21}. In contrast, the delay time τ_{21} becomes important in the non-Markovian dephasing, since the time-correlated frequency fluctuation depends on the first and second excitations within the initial dephasing process. Therefore, non-Markovian behavior can be observed in the SWM measurement as a function of the delay time τ_{21}, with a fixed delay time τ_{32}. The intensity change in the Markovian dephasing is eliminated by fixing the delay time τ_{32}, even if the QD ensemble shows the inhomogeneity of T_2.

The experimental result of SWM measurement at 3.4 K is shown in Fig. 2(b). The second delay time τ_{32} was set to 50 ps. The SWM signal clearly depends on the delay time τ_{21}, while it does not depend on τ_{21} in the Markovian approximation. The experimental result is composed of two coherent dynamics: the beat signal within 10 ps and the gradual decrease within 20 ps. The beat signal is caused by the interference between the exciton and biexciton transitions [30,31]. This exciton-biexciton beat is explained in the Markovian dynamics. In contrast, the gradual decrease within 20 ps corresponds to the non-Markovian dynamics due to the time-correlated frequency fluctuation of the exciton resonance energy. The correlation in the QDs is

maintained until 20 ps. The measured correlation time is one order of magnitude longer than the typical correlation time of excitons in semiconductors [11,12,15,18,19,27]. This is the first observation of long-time correlation over 10 ps in semiconductors using the time correlation measurement based on the SWM technique. This non-Markovian behavior was also observed for other delay times τ_{32}; the correlation time is independent of τ_{32}.

In order to understand the experimental result of the SWM measurement, we discuss the theory of the microscopic dephasing process. When there is no time correlation in the dephasing process, an excitonic polarization decays exponentially: $P(t) \propto e^{-t/T_2}$, which is the Markovian dynamics. In contrast, when the excitonic polarization maintains the time correlation, the non-Markovian dephasing takes place: $P(t) \propto e^{-S(t)}$, where $S(t)$ is a time-dependent function determined by the frequency fluctuation in the dephasing process, e.g., the exciton-phonon interaction. The time correlation has been investigated by using FWM measurements [9,14,16,18]. In these FWM measurements, the cumulant expansion method up to the second order of the interaction, leading to the dephasing, was used in the theoretical approach to the time correlation [6,7,18]. The SWM intensity is then calculated as follows:

$$I_{\mathrm{SWM}}(\tau_{32},\tau_{21}) \propto \Theta(\tau_{32}-\tau_{21})\Theta(\tau_{32})\Theta(\tau_{21}) \times \exp[-\{4S(\tau_{21})+8S(\tau_{32}) + 4S(\tau_{32}-\tau_{21})-4S(2\tau_{32}-\tau_{21}) - 4S(\tau_{32}+\tau_{21})+2S(2\tau_{32})\}], \quad (1)$$

where $\Theta(t)$ is a step function. The time-dependent function $S(t)$ is the time integral of the real part of the correlation function: $S(t)=\int_0^t dt_1 \int_0^{t_1} dt_2 \mathrm{Re}[\langle V(t_1)V(t_2)\rangle]/\hbar^2$, where $\langle V(t_1)V(t_2)\rangle$ is the correlation function of the interaction V leading to the frequency fluctuation [18]. This equation accurately expresses the gradual transition from non-Markovian to Markovian dephasing.

In the Markovian process, the correlation function $\langle V(t)V(0)\rangle$ is expressed by an instantaneous scattering, proportional to a delta function $\delta(t)$, which leads to $S(t)=t/T_2$. By substituting this time-dependent function into Eq. (1), the normal Markovian dephasing is obtained as the exponential decay model: $I_{\mathrm{SWM}}(\tau_{32},\tau_{21}) \propto e^{-4\tau_{32}/T_2}$. The theoretical SWM intensity in this T_2 model is shown by the solid line in Fig. 3(a). It shows the constant intensity as already discussed. In contrast, the SWM intensity depends on the delay time τ_{21} when the transient time correlation is taken into account. The calculated result for the simple stochastic model is shown by the dotted line in Fig. 3(a). In this calculation, the correlation function is assumed to be $\langle V(t)V(0)\rangle = \hbar^2 D^2 e^{-2t/\tau_c}$, where D and τ_c are the coupling constant and correlation time of the interactions, respectively. This stochastic model is widely used in atomic gases, liquids, and semiconductors [8,9,14–16]. The SWM intensity

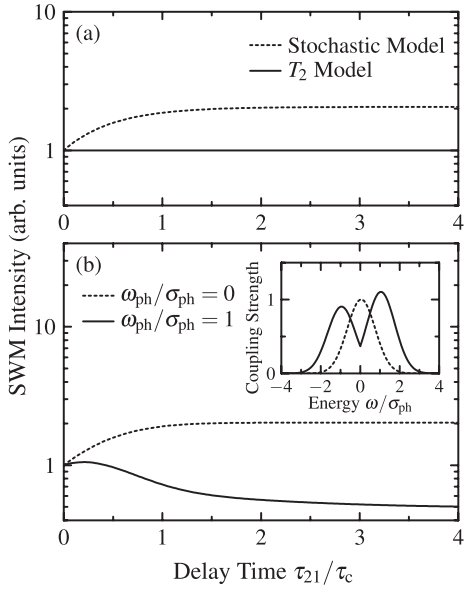

FIG. 3. Theoretical six-wave mixing intensity as a function of the normalized delay time τ_{21}/τ_c, where τ_c is the correlation time of each model. (a) Six-wave mixing intensities for the simple stochastic model (dotted line) and the T_2 model (solid line). (b) Six-wave mixing intensities for the microscopic phonon model with $\omega_{\mathrm{ph}}/\sigma_{\mathrm{ph}}=0$ (dotted line) and 1 (solid line). The inset shows the coupling strength of phonons corresponding to $\omega_{\mathrm{ph}}/\sigma_{\mathrm{ph}}=0$ (dotted line) and 1 (solid line).

increases in the time range maintaining the correlation, i.e., for $\tau_{21}<\tau_c$. This shows that the time correlation is an important characteristic of the frequency fluctuation for understanding the non-Markovian dynamics. However, the simple stochastic model does not explain the decrease in the experimental result. The non-Markovian behavior is caused not by carrier-dependent processes, e.g., exciton-exciton interactions, but by exciton-phonon interactions, since the dephasing constant T_2 does not show the excitation intensity dependence, but depends strongly on temperature. To explain the non-Markovian behavior of excitons in QDs, it is necessary to take into account the exciton-phonon interaction. The SWM intensity for the microscopic phonon model is shown in Fig. 3(b), where the correlation function is assumed to be $\langle V(t)V(0)\rangle = \int d\omega I_{\text{ex-ph}}(\omega)\times\{(n(\omega)+1)e^{-i\omega t}+n(\omega)e^{i\omega t}\}$. Here $n(\omega)$ is the Bose-Einstein distribution function of the phonon mode at a temperature T; i.e., $1/(e^{\hbar\omega/k_B T}-1)$, and $I_{\text{ex-ph}}(\omega)$ is the spectral density of the exciton-phonon interaction [6,17,18]. In bulk semiconductors, the spectral density of acoustic phonons is determined by phonon dispersion. For semiconductor QDs, the phonon coupling is determined by the QD confinement potential and the coupling to the characteristic phonon mode ω_{ph} enhanced by the excitonic energy structure. In the calculation, the spectral density with the center frequency ω_{ph} is assumed to be a Gaussian distribution $\alpha^2\hbar^2\omega e^{-(\omega-\omega_{\mathrm{ph}})^2/\sigma_{\mathrm{ph}}^2}$, where α is a dimensionless coupling

constant and σ_{ph} is the linewidth. The correlation time of the interaction, τ_c, is expressed by $2/\sigma_{ph}$. The coupling strength of the phonons, i.e., the Fourier transform of the correlation function $\langle V(t)V(0)\rangle$, is shown in the inset of Fig. 3(b). The calculated result for the centered phonon coupling $\omega_{ph} = 0$ is shown by the dotted line. The SWM intensity shows an increasing tendency that is similar to the result in the simple stochastic model shown in Fig. 3(a), which differs from the decrease in the experimental result. The decrease in the SWM intensity can be explained by the phonon coupling with $\omega_{ph} \neq 0$ as shown by the solid line in Fig. 3(b). This calculation indicates that the excitons in the QDs are strongly coupled to spectrally split phonon modes.

To understand the details of the non-Markovian dephasing, we quantitatively compared the calculation with the experimental result by taking into account the QD properties. The exciton states are split into X and Y exciton states due to the anisotropy of the QD confinement potential. This is known as the fine-structure splitting of exciton states [30,31]. The phonon coupling is enhanced at a phonon energy resonant with this fine-structure splitting, since the virtual transition between these exciton states is caused by the exciton-phonon interaction. The theoretical SWM intensity was calculated as shown in Fig. 4. In the calculation, the center value of the phonon coupling energy $\hbar\omega_{ph}$ was set to 87 μeV, which is the fine-structure splitting energy measured in our previous study [30]. The inhomogeneous broadening of the splitting energy in the QD ensemble was also taken into account. The theoretical result with the parameters $\alpha = 0.15$ and $\sigma_{ph} = 0.7\omega_{ph}$, shown by the solid line in Fig. 4, reproduces well the experimental result shown in Fig. 2(b). The exciton-biexciton beat was calculated independently of the dephasing process as discussed

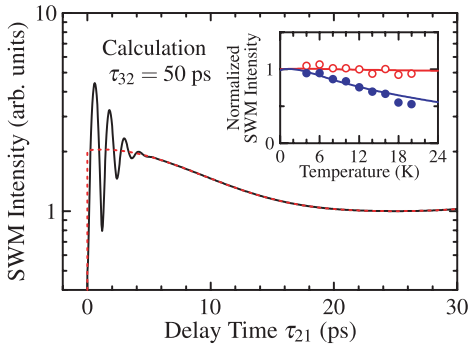

FIG. 4 (color online). Theoretical six-wave mixing intensity for the first delay times of $\tau_{21} = 0$ to 30 ps with (black solid line) and without (red dotted line) the exciton-biexciton beat. The second delay time τ_{32} was set to 50 ps. The inset shows the experimental (circles) and theoretical (lines) temperature dependence of the normalized six-wave mixing intensity. The second delay time τ_{32} was set to 40 ps. The experimental results are plotted at the delay times $\tau_{21} = 0.9$ (open circle) and 3.9 (solid circle) ps. The intensities are normalized by the intensity at the delay time of 0.3 ps to clarify the non-Markovian behavior.

in our previous study [30,32]. The non-Markovian behavior without the exciton-biexciton beat is shown by the dotted line. It clearly shows the decrease in the SWM intensity due to non-Markovian dynamics. The correlation time τ_c was found to be 23 ps from the definition of $2/\sigma_{ph}$. The long-time correlation is caused by the modulation of the spectral density of the exciton-phonon interaction. The QD confinement makes it possible for excitons to couple to the broad phonon spectrum due to the deformation potential coupling. In this broad spectrum, the total coupling strength is determined by the excitonic energy structure, i.e., phonon creation and annihilation due to virtual transitions. The phonon coupling in the QDs is concentrated at the resonant phonon mode; the narrow spectral density causes the long correlation. In the previous investigation of phonon coupling in QDs [10,11], the initial decoherence was caused by a broad phonon coupling due to the exciton confinement. This broad coupling does not explain the long-time correlation in our study. The coupling spectrum should be one order of magnitude narrower than the spectrum due to the confinement. The narrow spectrum is strongly enhanced by the virtual transition resonant with the fine-structure splitting in the strain-controlled QDs grown on the (311) plane, which are different from the usual QDs grown on the (100) plane [30,31].

We measured the temperature dependence of the SWM intensity to understand the phonon density dependence. The experimental temperature dependence is plotted in the inset of Fig. 4. To eliminate the influence of the exciton-biexciton beat, the SWM intensity was measured at the nodes of the beat: $\tau_{21} = 0.9$ and 3.9 ps. This temperature dependence clearly captures the non-Markovian dynamics depending on the phonon density. The SWM intensity for $\tau_{21} = 3.9$ ps shows the gradual decrease with temperature, but the intensity for $\tau_{21} = 0.9$ ps maintains the coherence. This persistence is the result of the SWM decoherence suppression; i.e., it is a reversing technique for the dephasing process [27]. The calculated results, shown by the lines in the inset, correspond well to the experimental results. The time-correlated frequency fluctuation due to the exciton-phonon interaction is accurately explained by the microscopic model, taking into account the phonon coupling in QDs.

In summary, we have investigated time correlation in the non-Markovian dephasing process for InAs quantum dots by using a six-wave mixing technique. Since the time correlation measurement based on this technique is free from the influence of Markovian dephasing, a pure time correlation of the frequency fluctuation can be obtained in the measurement. For the first time, a long-time correlation over 10 ps has been observed by using the time correlation measurement, which is one order of magnitude longer than conventional correlation times. The non-Markovian behavior is successfully explained by a microscopic dephasing model that takes into account the frequency fluctuation

due to exciton-phonon interactions. We have clarified that the long-time correlation is caused by the modification of phonon coupling distribution in quantum dots. The time correlation measurement presented in this study is a powerful tool to analyze the initial non-Markovian dephasing. It will provide valuable information about the underlying nature of time-correlated dynamics for other quantum dynamical systems.

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