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Effects of pumping on propagation velocities of confined exciton polaritons in GaAs/$Al_xGa_{1-x}As$ double heterostructure thin films under resonant and non-resonant probe conditions

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We report the effects of excitation of exciton polaritons on their propagation in GaAs (110 nm)/$Al_{0.3}Ga_{0.7}As$ double heterostructure thin films by measuring the cross-correlation signal, which was recorded as the intensity of the second harmonic light generated by the gate pulse and the probe pulse reflected from the sample. When the probe energy is tuned at the lowest exciton energy, the signal profile changes due to the appearance of nonlinear dispersion. On the other hand, the signal profile shows a complicated change under the non-resonant probe condition, in which the energy is the center energy of two exciton states. These results originate from a change in the propagation velocity of the exciton polariton due to the pump. © *2013 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4772717]

I. INTRODUCTION

The propagation of a light pulse through dispersive materials is important in ultrafast physics because of various interesting phenomena including self-induced transparency,[1,2] electromagnetic-induced transparency,[3,4] polariton beats,[5–8] and slow light in nanostructured semiconductors.[9–13] Moreover, semiconductor microcavity structures and waveguides, which thicknesses are comparable to or larger than the light wavelength, are used to control the interaction between an exciton and a light field.[14–19] When the frequency of the light pulse corresponds to the exciton energy, the light pulse propagates as an exciton polariton. The propagation of the exciton polariton is characterized by a group velocity, which is much slower than light velocity. This slow propagation induces interesting physics such as slow light.[9–13] These phenomena are essential for the creation of new communication technologies such as quantum information and ultrafast optical communications.

We recently reported the propagation of an exciton polariton confined in GaAs/$Al_{0.3}Ga_{0.7}As$ double heterostructure thin films by using a transient-grating and a pump-probe techniques.[20–24] In these measurements, the signals showed rise components at the exciton energies in the negative time region. The velocities evaluated from the rise time and the film thickness correspond to the group velocities at the exciton energies in the bulk GaAs crystal. These results also demonstrate that one of the factors limiting the exciton polariton coherence time is the propagation (group) velocity. Moreover, an ultrafast response due to exciton quantum beats was observed by a degenerate four-wave-mixing[25,26] and a pump-probe techniques.[24] The fast relaxation observed by the pump-probe technique represents population modulation, which is useful for the application of optically controlled ultrafast devices.

In this work, we investigated the change in propagation characteristics of the exciton polariton due to the pump by measuring the cross-correlation signal. Because our sample size is approximately equal to half of the light wavelength, the exciton-polariton mode coherently extends over the entire sample along the growth direction, which enhances the coupling strength of the light and exciton.[27,28] The cross-correlation signals generated by the probe and gate pulses show the change in the probe pulse profile caused by the pump of the exciton polaritons. The change cannot be explained by the reflectivity change corresponding to population modulation. Furthermore, the change varies with detection energy. The change in the characteristics of the propagating exciton polariton induced by the pump is discussed in terms of the change in the propagation velocity.

II. EXPERIMENT

The sample used in this study is the double heterostructure thin films with three periods of GaAs (110 nm)/$Al_{0.3}Ga_{0.7}As$ (5 nm) on a (001)GaAs substrate grown by molecular beam epitaxy. The GaAs layer thickness is larger than the exciton Bohr radius of 11 nm; the center-of-mass motion of the excitons is confined.[22,25] The transient response was measured using a reflection-type pump-probe technique. The light source used was a mode-locked Ti:sapphire pulse laser with a pulse width of approximately 200 fs and repetition rate of 80 MHz. The pump energy was tuned at the lowest exciton of 1.5158 eV. We varied the spectrum of the probe pulse by using a slit placed between a grating pair as shown in Fig. 1.[29] The spectral width ΔE of the probe pulses was reduced to 1.0 meV by changing the slit width, and the probe energy was tuned at 1.5158 eV and 1.5166 eV by changing the slit position. At the energy of 1.5166 eV, an ultrafast response due to a combination of quantum beats and momentum relaxation was observed.[24] The pump power was

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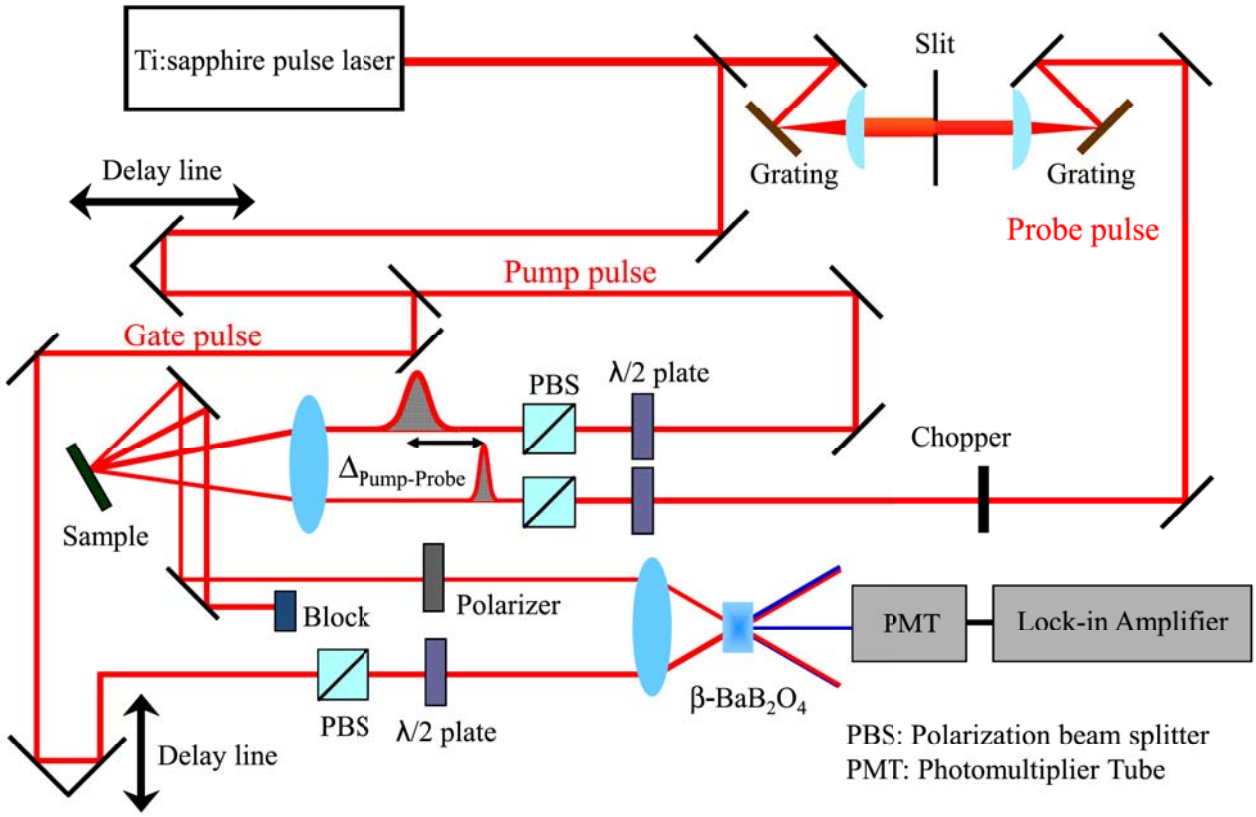

FIG. 1. Schematic diagram of cross-correlation measurement system. A slit and grating pair are used to control the spectrum of the probe beam. The intensity of the SH light generated by the probe beam reflected from the sample and the gate pulse was recorded as a function of time separation of those pulses under various $\Delta_{Pump\text{-}Probe}$.

changed from 0.1 to 0.3 mW, which corresponds to a change from 12 to 36 nJ/cm^2, while the probe power was kept at 0.01 mW. The cross-correlation signal, which corresponds to the temporal profile of the probe pulse reflected from the sample with or without the pump, was measured by generating the second-harmonic (SH) light with use of a $\beta-BaB_2O_4$ crystal. Figure 1 shows a schematic diagram of the cross-correlation measurement system. The SH light intensity was recorded as a function of the difference in the optical path length between the probe and gate pulses under various time delays between the pump and probe pulses $\Delta_{Pump\text{-}Probe}$. The intensity of the SH light was measured using a photomultiplier tube (PMT). The signal from the PMT was amplified by a lock-in amplifier. All measurements were performed at 4 K.

III. RESULTS AND DISCUSSION

First, we show the time-resolved reflectivity-change signals obtained by changing the pump power to demonstrate the transient characteristics of the exciton polariton in Fig. 2. The ΔE values for the pump and probe pulses were 20 meV and 1.0 meV, respectively. The signals show a rise component in the negative time region, and the rising time is independent of the pump power. The signal in the negative time region originates from the propagation of the exciton polariton confined in the thin films.[21–24] A positive reflectivity change indicates that the signal originates from the Coulomb-induced correlations.[30] The pump-probe signal shape hardly changes with the pump power, which indicates that the contribution of many-body effects caused by increasing the pump power is negligible in this pump power region.

Figure 3 shows the cross-correlation signals at $\Delta_{Pump\text{-}Probe} = 0$ ps. The dotted and solid curves indicate the signals measured in the pump-off and pump-on conditions, respectively. The decrease in the signal intensity mostly

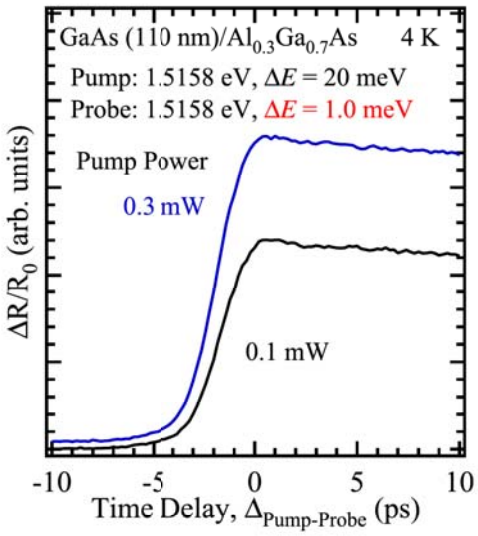

FIG. 2. Pump-probe (time-resolved reflectivity change) signals obtained by changing the pump power in the sample at 4 K under the exciton resonance condition. The probe ΔE was 1.0 meV.

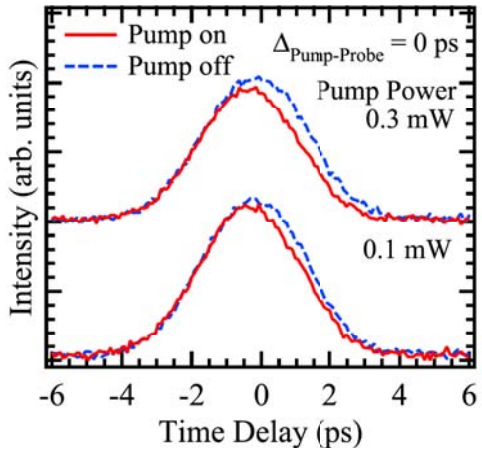

FIG. 3. Cross-correlation signals measured under the pump-probe conditions of Fig. 2. $\Delta_{\text{Pump-Probe}}$ was kept at 0 ps. The dotted and solid curves indicate the SH intensities measured in the pump-off and pump-on conditions, respectively.

appears in the positive time region. Because $\Delta_{\text{Pump-Probe}} = 0$, the change in the intensity originates from the generation of the exciton polariton by the pump pulse. However, if this change is caused by the reflectivity change shown in Fig. 2, the SH intensity should be increased by the pump; i.e., the SH intensity decreases not due to the reflectivity change but due to the direct modulation of the pulse shape. This is a remarkable point in this work.

To elucidate the origin of the modification of the SH signal profile, we measured the correlation signal with various $\Delta_{\text{Pump-Probe}}$ values, as shown in Fig. 4. At the negative $\Delta_{\text{Pump-Probe}}$, there is no change in the correlation signal due to the pump. A negative $\Delta_{\text{Pump-Probe}}$ means that the probe pulse reaches the sample before the pump pulse. Hence, the modulation of the correlation signal is caused by the excitation of the exciton polariton.

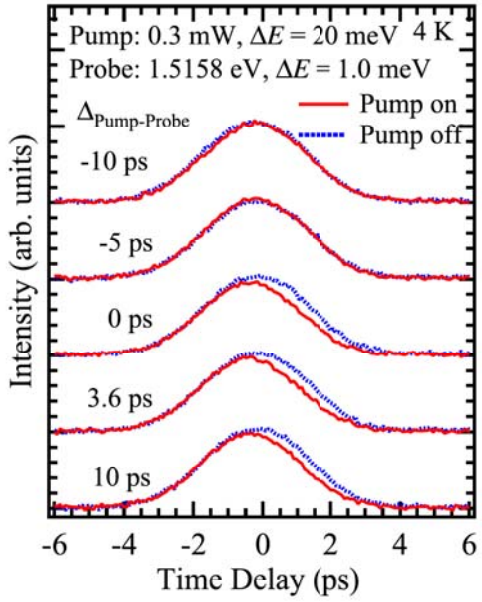

FIG. 4. Cross-correlation signals measured under a pump power of 0.3 mW at various $\Delta_{\text{Pump-Probe}}$ values. The dotted and solid curves indicate the SH intensities measured in the pump-off and pump-on conditions, respectively.

On the other hand, at a positive $\Delta_{\text{Pump-Probe}}$, the correlation signals change in the positive time region, and the magnitude of the change slightly decreases with increasing $\Delta_{\text{Pump-Probe}}$. These tendencies measured at negative and positive $\Delta_{\text{Pump-Probe}}$ values do not depend on the pump power in this power region. A positive $\Delta_{\text{Pump-Probe}}$ means that the probe pulse reaches the sample after the pump pulse. Therefore, these results support our suggestion that the change is related not to the modulation of the exciton population but to direct modulation of the pulse shape of the exciton polariton. Furthermore, the fact that the SH signals change only in the positive time region is noteworthy. For instance, at $\Delta_{\text{Pump-Probe}} = 10$ ps, the probe pulse passes through the sample 10 ps after the exciton polariton is generated. Nevertheless, the front portion of the pulse does not change, and only the rear portion is modulated. These features observed under positive and negative $\Delta_{\text{Pump-Probe}}$ values are inconsistent with the possibility that the change originates in modulation by the exciton polariton.[6,7]

To clarify the amount of change in the signal intensity, we evaluated the ratio of the signal intensities, $(I_{\text{off}} - I_{\text{on}})/I_{\text{off}}$, where I_{on} and I_{off} indicate the signal intensities with and without the pump, respectively. In Figs. 5(a) and 5(b), the evaluated results are summarized for an effective time region from −2.0 ps to 2.5 ps. The existence of the time delay dependence clearly disproves population modulation as the origin of the SH signal change, because the exciton lifetime is approximately 1.5 ns.[2] In Fig. 5(a), which shows the pump power dependence at $\Delta_{\text{Pump-Probe}} = 0$ ps, the amount of change in the SH intensity depends on the pump power; the ratio increases with increasing pump power. On the other hand, in Fig. 5(b), which shows the $\Delta_{\text{Pump-Probe}}$ dependence

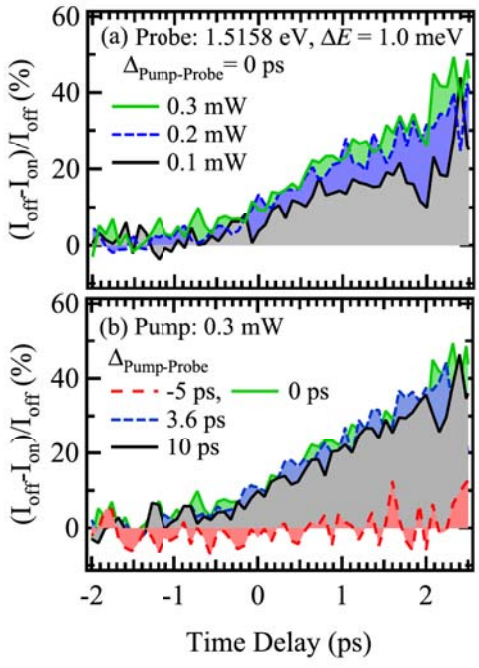

FIG. 5. Ratios between the SH signals measured with and without the pump. The signals obtained by (a) changing the pump power at $\Delta_{\text{Pump-Probe}} = 0$ ps and (b) changing $\Delta_{\text{Pump-Probe}}$ at a pump power of 0.3 mW (b) were evaluated.

at a pump power of 0.3 mW, the ratio decreases as $\Delta_{\text{Pump-Probe}}$ increases. Because an increase in $\Delta_{\text{Pump-Probe}}$ corresponds to the dephasing of the exciton polariton, this result demonstrates that the coherence of the exciton polariton generated by the pump is closely related to the modulation of the propagation pulse.

A decrease in the SH intensity observed in the positive time region corresponds to an increase in the propagation velocity. The increase in the propagation velocity with increasing pump power disproves the existence of the slow light effect which reduces the velocity as the power increases.[9] Moreover, on the basis of the spectral measurements, we have reported that the exciton peaks are not shifted by the pump.[21,23] Therefore, the change in the propagation velocity does not originate from self-induced transparency.[2]

As a possible cause, we considered the change in the dispersion induced by the pump.[31] An excitation by the broadband pump pulse changes the polariton dispersion curve and produces a nonlinear dispersion term, which consequently modulates the propagation velocity of the exciton polariton. This modulation depends on the time delay between the probe and gate pulses; the traveling process in the sample enhances the increase in the velocity. Furthermore, the existence of the $\Delta_{\text{Pump-Probe}}$ dependence indicates that the coherence of the exciton polariton generated by the pump pulse is an important factor in the propagation velocity change. Unfortunately, the theoretical reason that the front portion is not modulated is unclear. We suppose that the front part may propagate as a soliton-like pulse or that the time resolution of the system limits the observation of the change. These results indicate the importance of considering the nonlinear dispersion term even in confined states.

Next, we demonstrate the results under another probe condition to show the effect of the ultrafast relaxation on the propagation characteristics. Figure 6 shows the reflectivity change signals probed at 1.5166 eV. At this energy, the ultrafast response is caused by a combination of quantum beats and momentum relaxation.[24] Because the fast response ends at around 3.6 ps, hereafter, we focus on the phenomena at this time delay.

Figure 7 shows the cross-correlation signals measured at various $\Delta_{\text{Pump-Probe}}$ values under the pump-on and pump-off conditions, which are depicted by the solid and dotted curves, respectively. In the negative $\Delta_{\text{Pump-Probe}}$ region, the SH signal hardly changes by the pump, and the front portion of the pulse in all $\Delta_{\text{Pump-Probe}}$ regions is not modulated. These results are the same as those under a probe with an energy of 1.5158 eV. An apparent difference is the intensity increase due to the pump. Furthermore, an intensity inversion occurs at around 3 ps; the SH intensity decreases after 3 ps. Namely, when $\Delta_{\text{Pump-Probe}} > 3.6$ ps, the propagation velocity decreases for 3 ps and then increases after 3 ps.

Although the SH signal intensity hardly changes at $\Delta_{\text{Pump-Probe}} = 0$ ps, the intensity increases at $\Delta_{\text{Pump-Probe}} > 3.6$ ps. This probe energy of 1.5166 eV is not for the specific exciton state but for the center energy between the $n = 2$ light hole (1.5164 eV) and $n = 4$ heavy hole (1.5168 eV) excitons.[24] Therefore, during the excitons maintain their coherence, the light pulse can travel in the sample as in a transparent material.

To clarify the amount of change in the signal intensity, we evaluated the ratio of the signal intensities as described above. Figure 8 summarizes the results for an effective time region from −1.5 ps to 5 ps. The ratio increases when $\Delta_{\text{Pump-Probe}} > 3.6$ ps. Furthermore, the ratio reaches a peak at around 2 ps, and the sign changes from negative to positive between 3 and 4 ps. These phenomena have not been observed under a probe energy of 1.5158 eV.

The appearance of the peak around 2 ps indicates that the propagation velocity reaches a minimum at around 2 ps and is then accelerated. In the transient signals shown in Fig. 6,

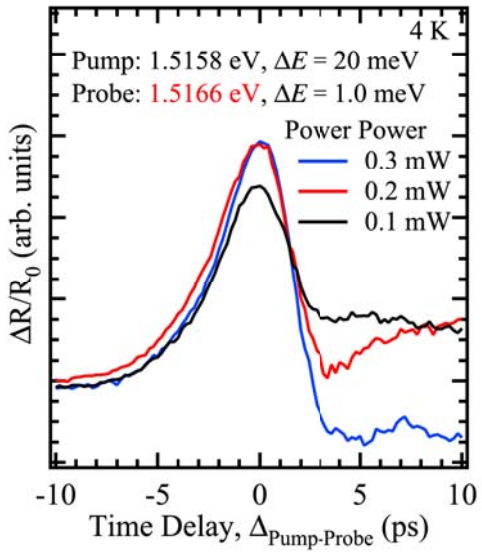

FIG. 6. Pump-probe signals obtained by changing the pump power in the sample at 4 K probed at 1.5166 eV. The probe ΔE was 1.0 meV.

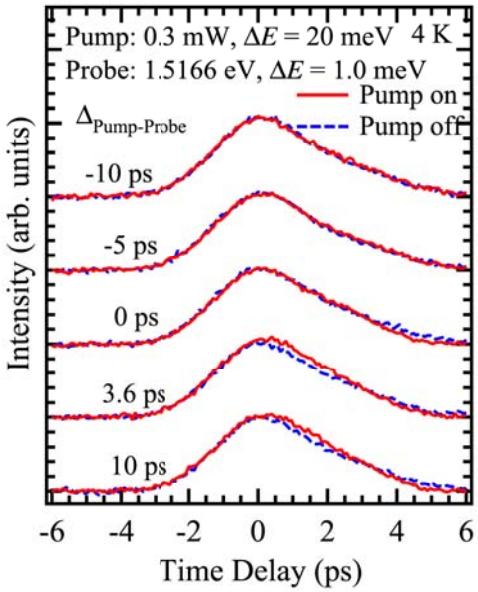

FIG. 7. Cross-correlation signals measured under the pump power of 0.3 mW at various $\Delta_{\text{Pump-Probe}}$ values for a probe energy of 1.5166 eV, which causes ultrafast relaxation. The dotted and solid curves indicate the SH intensity measured in the pump-off and pump-on conditions, respectively.

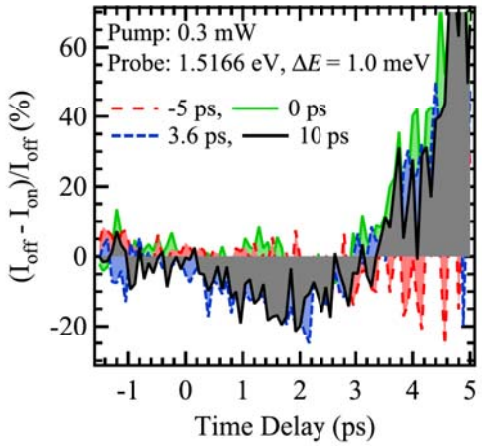

FIG. 8. Ratios between SH signals with and without the pump. The signals obtained by changing $\Delta_{\text{Pump-Probe}}$ at a pump power of 0.3 mW were evaluated.

the quantum beat oscillation ends at $\Delta_{\text{Pump-Probe}} = 3.6$ ps. Therefore, this acceleration and reduction in velocity do not depend on the exciton coherence. Moreover, if the exciton population generated by the pump pulse causes the SH signal modulation, the results in the negative time region at $\Delta_{\text{Pump-Probe}} = 3.6$ ps and 10 ps cannot be explained. Thus, we measured the pump power dependence at $\Delta_{\text{Pump-Probe}} = 3.6$ ps.

Figure 9 summarizes the ratios between the SH signals with and without the pump obtained by changing the pump power at $\Delta_{\text{Pump-Probe}} = 3.6$ ps. The ratio hardly changes with the pump power, which cannot be explained by the slow light effect and the appearance of nonlinear dispersion.

One possible reason is the quantum beat oscillation. At $\Delta_{\text{Pump-Probe}} = 3.6$ ps, the $n = 4$ heavy hole exciton, which is the upper state of the quantum beat, dephases because the dephasing of the quantum beat oscillation observed by the pump-probe technique originates from the exciton dephasing due to an intersubband transition.[32,33] This dephasing accelerates the polariton. In addition, the propagating pulse is also affected by the quantum beat and corresponds to the phase of the quantum beat oscillation.[6,7] Therefore, the dephasing of

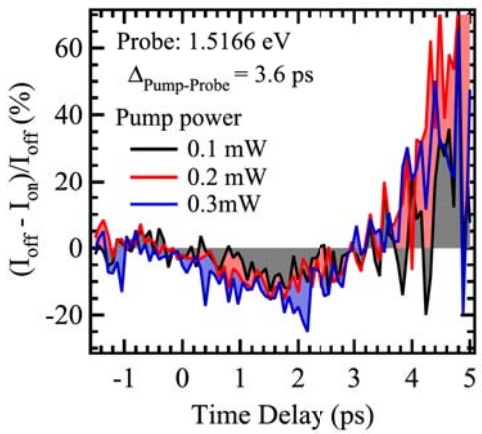

FIG. 9. Ratios between SH signals with and without the pump. The signals obtained by changing the pump power at $\Delta_{\text{Pump-Probe}} = 3.6$ ps were evaluated.

the $n = 4$ heavy hole exciton increases the velocity. That is, in the quantum beat condition, the velocity is determined by the balance between the polariton beating and exciton dephasing.

IV. CONCLUSION

We investigated the effect of excitation of the exciton polariton on its propagation in GaAs (110 nm)/$Al_{0.3}Ga_{0.7}As$ double heterostructure thin films by measuring the cross-correlation signal, which was recorded as the intensity of the SH light generated by the gate pulse and the probe pulse reflected from the sample. When the probe energy is tuned at the exciton energy, the change in the SH signal profile due to the pumping of the exciton does not correspond to that in the reflectivity change. Although the reflectivity change originates from a change in the refractive index, the change in the SH signal profile results from the appearance of nonlinear dispersion. On the other hand, the behavior of the SH signal profile varies with the probe energy. The SH signal profile shows a complicated change under the non-resonant probe condition at which the energy is the center energy between those of the two exciton states. In this case, the quantum beat affects the change in the signal profile. These interesting results indicate that a strict control of the probe pulse will clarify the propagation characteristics of the exciton polariton, which is an important insight for creating devices such as optical delay devices.

ACKNOWLEDGMENTS

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