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Picosecond time Resolved Spectra of the Optical Stark Effects on the Stacking Fault Excitons in BiI_3 Crystals

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PICOSECOND TIME RESOLVED SPECTRA OF THE OPTICAL STARK EFFECTS ON THE STACKING FAULT EXCITONS IN BiI_3 CRYSTALS

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Abstract We report the picosecond time resolved spectra of the optical Stark effect on a quasi-two-dimensional exciton localized at a stacking fault plane in BiI_3 crystals. The ultrafast response of the blue-shift due to the optical Stark effect(OSE) is confirmed in picosecond time domain under negative detuning excitation condition. For positive detuning excitation, an ultrafast switching from the red shift by the OSE to the blue-shift due to the nonlinear interaction with the large amount of incoherent "real" exciton occurs. Under the just resonance excitation, the blue-shift due to the "real" exciton is also observed during several tens of picoseconds after the pumping, and an absorption bleaching occurs at the timing of the pump-pulse peak. The bleaching is discussed in connection with the OSE.

INTRODUCTION

In BiI_3 crystals, a favorable exciton system exists for investigation of nonlinear effects of exciton. That is a localized exciton system at a stacking fault plane produced accidentally during the crystal growth. We call these excitons stacking fault excitons (SFE's).[1] A typical absorption spectrum is shown in Fig.1-(a). The R-, S-, and T-lines originates from the SFE transitions. The SFE's have very sharp absorption profiles. The delayed photon-echo measurements have shown their long coherence time of several tens of picoseconds.[2] These characteristics of the SFE's allow us to observe spectral changes due to the intense laser field providing a good tool to study the optical Stark effect (OSE) on exciton systems.

By intense laser pumping, various spectral changes are induced. The quantities of the spectral changes depend drastically on the excitation laser frequency $\hbar\omega_L$. Figure 1-(b) shows the $\hbar\omega_L$ dependences of the peak-shift values of the R-, S- and T-lines. The resonant enhancement of peak-shift and the discontinuous peak-shift jump on each line take place for the $\hbar\omega_L$ around and at all SFE resonances.[3] A simplified "dressed" exciton model[4] expanded to a composite four-level system has derived the consistent results[3] to the observed $\hbar\omega_L$ dependences of the peak-shift except for an additional blue-shift due to a large amount of really excited excitons.[5,6]

In this paper, the ultrafast response of the OSE in picosecond time domain are reported in the SFE system.

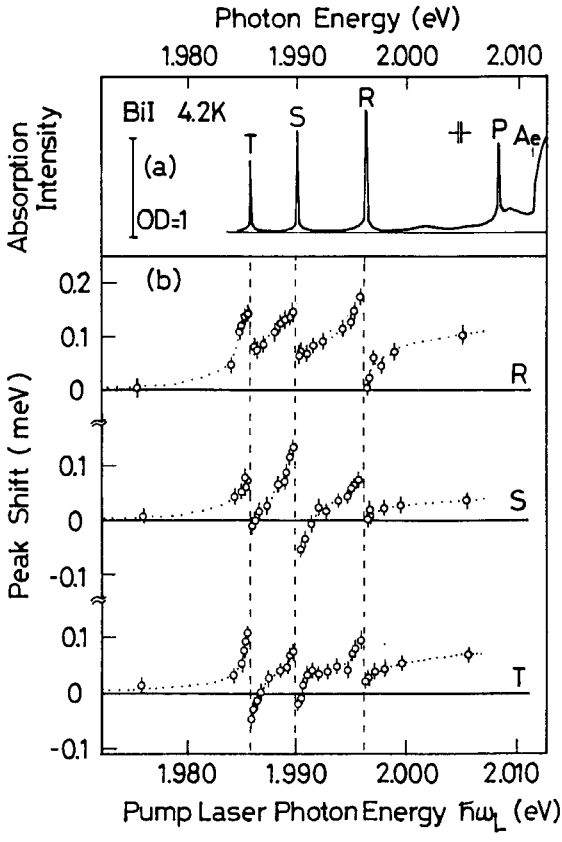

FIGURE 1 **(a)** A typical absorption spectrum of BiI_3. R, S, and T are due to the SFE's, A_e: the absorption step due to the indirect exciton assisted with the phonon A emission. P is another origin. **(b)** $\hbar\omega_L$ dependences of peak-shift values of the R-, S-, and T-line. Vertical broken lines denote the respective resonant energies of the R, S, and T.

EXPERIMENTAL

We employed the pump-and-probe method shown in Fig.2. A cavity dumped dye laser pumped by a mode-locked CW Nd:YAG LASER was used for the light-source. The pulse duration and the spectral width of the laser light were $\sim$ 3psec and $\sim$ 2meV

in FWHM, respectively. The excitation density of pump-pulse is $\sim 10^5 W/cm^2$. The spectral width is wide enough to probe the SFE lines. The weakened probe light gives transient absorption spectra by simultaneous measurements of the probe beam reference.

RESULTS and DISCUSSION

Figure 3 shows the delay-time dependence of the absorption peak energies for several values of detuning parameter $\Delta\hbar\omega_L^T$. For the negative detuning excitation, the time response of the peak-shift (○) follows the excitation laser field, i.e. the blue-shift of the T-line increases with rising pump-pulse and decreases immediately with decreasing pump-pulse. This result confirms the ultrafast response shorter than 2psec of the OSE on the SFE.

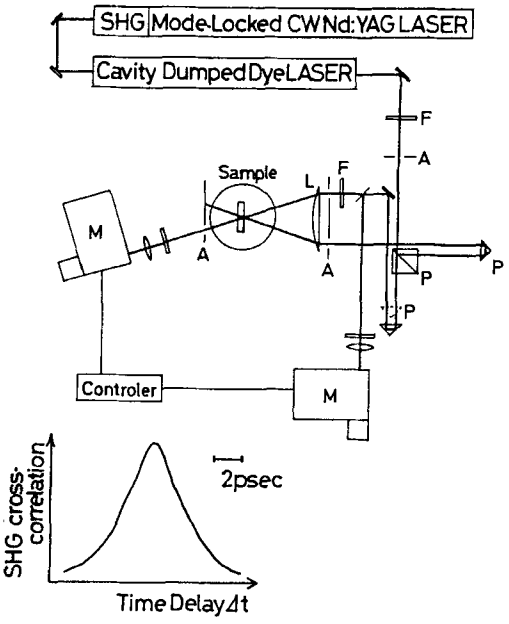

FIGURE 2 Experimental setup for measuring the time-resolved spectra in the picosecond time domain. F:filter, A:aperture, L:lens, P:prism, M:monochromator. The inset shows the SHG cross-correlation trace.

Under the positive detuning excitation, on the other hand, the peak-shift values show a peculiar Δt-dependence ($\triangle, \triangle$). The T-line peak shifts slightly to be red at the very moment the pump-pulse stands up ($\Delta t \sim$ 2psec), and the shift changes its sign with increasing Δt. The blue-shift grows until the end of the pump-pulse and keeps its value after the fading out of the pump-pulse. The red-shift is expected for the positive detuning from the simplified "dressed" exciton model.[3] In general, the spectral dispersion of the ultrashort pulse makes it hard to tune finely to the exciton resonance. Even for a small detuning, the pump-pulse covers the exciton resonance, and the "real" excitons created by the overlapping of pump-laser obscure the

coherent nature of "virtual"[7] exciton which were to be excited by off-resonance excitation. Under our experimental condition, $\lesssim$10% of the pump-pulse intensity is absorbed by the T exciton. From the population decay time constant $\sim$ 1nsec of the T exciton[2], the population of the "real" exciton can be roughly estimated to be $\sim 10^{10\pm1}/\mathrm{cm}^2$ at the timing of pump-pulse peak. The high density exciton should induce the nonlinear interaction between the exciton created by the probe-light and the incoherent "real" excitons, resulting in the blue shifts.[2]

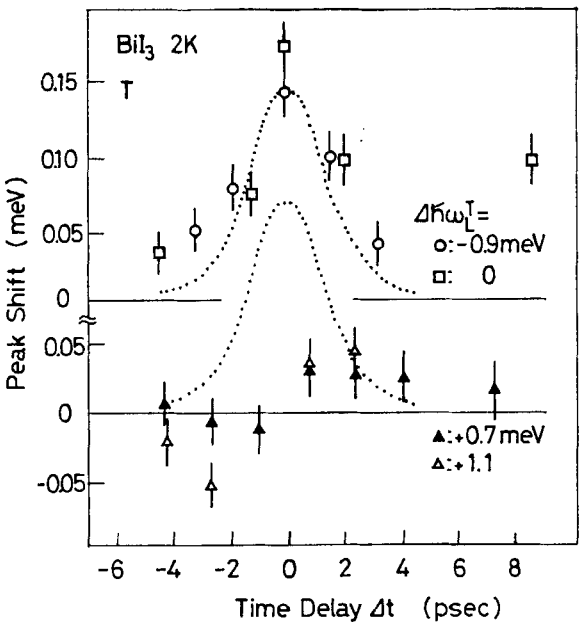

FIGURE 3 Delay-time dependence of the T-line peak-shift $\Delta\hbar\omega_T$ in the picosecond time domain; Δt is the time delay of the probe-pulse peak from the pump-pulse peak; $\Delta\hbar\omega_L^T$ are the detuning parameters of the peak energy of the pump-laser from the T resonance. The dotted line illustrates the temporal profile of the pump-pulse calculated by assuming the hyperbolic-secant function.

For the detuning of $\Delta\hbar\omega_L^T=0$, the similar blue-shifts to that under the negative detuning condition appear during the pump-pulse, and a finite blue-shift keeps its value for several tens of picoseconds as shown by □ in Fig.3. The residual blue-shift is also considered to be the nonlinear effect due to the high density "real" exciton. A bleaching of absorption occurs at $\Delta t=0$. Figure 4 shows the excitation intensity dependence of the T-line absorption at $\Delta t=0$. The blue-shift and the absorption bleaching on the low energy tail appear. The bleaching and the shift become more remarkable with increasing excitation intensity. Under the just resonance excitation, a large splitting of "dressed" exciton states and a significant absorption reduction[3,8,9] are expected

on account of an efficient coupling between the exciton and the intense laser field. Thus, it is suggested that the absorption bleaching at $\Delta t=0$ comes from the resonant OSE. The phenomena, however, are much complicated because of the real excitation of the large amount of excitons. In GaAs/AlGaAs multi quantum well, the ionization process of the resonantly excited exciton governs the dynamics of the absorption bleaching in ultrafast time domain.[10] On the contrary, the large amount of the "real" exciton only brings about the energy shift and broadening without bleaching on the SFE system in BiI_3.[11] This fact supports the interpretation of our system for the bleaching.

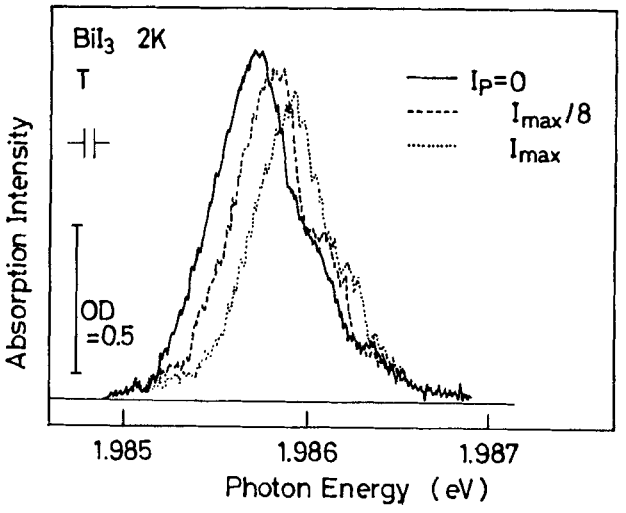

FIGURE 4 Excitation intensity dependence of the T-line absorption spectra. The solid line illustrates the regular absorption spectrum without pumping. The maximum excitation intensity $I_{max}=8 \times 10^5 W/cm^2$.

SUMMARY

The picosecond time-resolved absorption spectra of the optical Stark effects on the SFE system in layered crystal BiI_3 have been presented. Under negative detuning excitation condition, the ultrafast response of the optical Stark shift has been confirmed in picosecond time domain. Both the red shift due to the optical Stark effects and the blue shift due to the large amount of incoherent "real" exciton have been resolved under positive detuning excitation. Under just resonance excitation, it has been observed that the blue-shifts originating

from the nonlinear effect due to the "real" exciton keeps during several tens of picosecond, and the significant absorption bleaching appear at $\Delta t=0$.

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