Observation of optical Stark effect between 1s - 2p exciton levels in CuCl single crystal

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Abstract. We measured the transient reflection spectra induced by infrared light for a CuCl single crystal using pump-probe spectroscopy. A Stark shift of the 1s exciton was observed using light with a near resonant energy between the 1s and 2p exciton levels. The amount of energy shift due to the Stark effect was obtained from the absorption spectra calculated using the Kramers–Kronig transform. We investigated the energy shift of 1s excitons with respect to the excitation density. This is the first published report on the Stark effect of 1s exciton associated with the internal energy levels of the excitons in CuCl crystal.

1. Introduction
When high intensity light such as a laser beam is incident on a material sample, a dressed state is generated in which an electron level of an atom is strongly coupled with a photon. The dressed state can be observed as an energy shift by the optical Stark effect during light irradiation, and the magnitude of the shift is determined by the intensity of the incident light and the detuning of the energy. In semiconductors, an exciton dressed state can be generated as in the case of an atomic system, because excitons have hydrogen atom-like energy. To observe the optical Stark effect in semiconductors, non-resonant light is generally used to avoid the complication of the optical response associated with the incoherent effect of excitons or carriers excited in the case of resonant excitation [1,2]. In addition, the optical Stark effect related to the internal levels of the exciton is quite useful because of the coherent control of atom-like levels in solid-states that it affords. For Cu2O and GaAs quantum well, the optical Stark effect of 2p exciton has been reported for infrared light resonant for 1s–2p transition energy [3,4].

In this study, we focused on an exciton in CuCl single crystal. CuCl is a direct transition semiconductor, and the exciton with the lowest energy (named as Z3 exciton) has a small Bohr radius and a large exciton binding energy (197 meV [4]). The internal energy levels related to the Rydberg states were observed by the absorption spectrum and the photoluminescence (PL) excitation spectrum [5]. In addition, transient absorptions from 1s to 2p and 3p exciton states, respectively, were observed via pump-probe spectroscopy [6]. The transition energy from 1s to 2p states was 169.5 meV. For CuCl, the Stark shift of the exciton has been reported when the photon energy of the pump light was near resonant to the transition energy from an exciton to biexciton states [7]. However, the Stark shift induced by the pump light resonant to the energy between the 1s and 2p levels of the exciton has not been
reported. We observed the optical Stark effect by time-resolved reflection spectroscopy. An energy diagram of this effect which was measured in this study is shown in Figure 1.

A reduction of reflectance originated from the irradiation of the pump light. From the absorption spectrum obtained by Kramers-Kronig transform (K-K transform), a red shift of the peak energy was confirmed, in addition to a reduction of the absorption coefficient and broadening of the spectral width at the pump light irradiation.

2. Experiment setup and method
The CuCl single crystal was fabricated by a vapor phase growth method. From theoretical calculations of the reflection spectrum to fit the experimental results, the damping constant of the exciton 1s level and the exciton dead layer were determined to be $\Gamma_{1s} = 0.75$ meV and $l = 11$ nm, respectively. Transient changes in the reflection spectra were measured by pump-probe spectroscopy using dual optical parametric amplifiers (OPAs) pumped by a regeneratively amplified pulse from a mode-locked Ti:sapphire laser. Time width of correlation between the pulses from two OPAs was 1.5 ps. The probe pulse was obtained by self-phase modulation, using a YAG crystal, of sun-frequency light (wavelength of 500 nm) of a signal and pump light from an OPA. A bandpass filter was used to select the wavelength near the 1s exciton energy in CuCl. The pump pulse in the mid-infrared region was obtained by difference frequency generation, using an AgGaS$_2$ crystal, of the signal and idler light from the other OPA.

The pump energy was tuned to 150 meV, 165 meV and 175 meV. The sample was positioned in a holder in a cryostat and temperature was kept at 3 K. The spot size of the pump light was 221 $\mu$m and that of the probe pulse was 215 $\mu$m in diameter on the sample surface. The relative delay times for the two pulses was controlled by a mechanical delay line. The reflection spectra were measured using a spectrometer (focal length: 500 mm; grating: 2400 g/mm; spectral resolution: 0.3 meV) equipped with a charge-coupled device (CCD).

3. Results and discussions
Figure 2 shows the contour image of the time-resolved reflection spectra at the excitation energy of 165 meV. A change in the reflectance was observed at the delay time of 0 ps. It is concluded that this is due to the Stark effect because the time width of the change in the reflectance is approximately equal to the pump pulse width. Figure 3 (a) shows the reflection spectrum (blue curve) at the time origin and that at $-65$ ps (black curve) without the influence of the pump light. It was confirmed that the reflectance decreased due to irradiation of the pump light.

In order to investigate the energy shift caused by the Stark effect in detail, the reflection
spectra were converted into absorption spectra using the Kramers-Kronig (K-K) transform as shown in Figure 3 (b). The blue and black solid curves are the absorption spectra at the time origin and the −65 ps, respectively. We observed a reduction of the absorption coefficient and a broadening of the spectral width. Figure 4 shows the changes in the absorption spectra at pump energies of 150, 165, and 175 meV (detuning of 19.5, 4.5 and −5.5 meV) with a pump density of 254, 366 and 315 μJ/cm², respectively. The positive peaks at 3.201 eV implies a red-shift of the 1s exciton energy level. In contrast, the negative change in the absorption from 3.202 to 3.210 eV was difficult to analyze. It is suggested that the complexity of this change is due to the crystallinity of the sample, e.g. the damping constant and the thickness of the dead layer. As a result of the Stark effect, the exciton energy splits into two levels (upper and lower levels). The red-shift shown in Figure 4 indicates the appearance of the lower levels. Simultaneously, the blue shift which originates from the upper level is expected. The oscillator strengths from the ground state to the lower and upper levels depend on the detuning energy; the oscillator strength of the lower level becomes strong for positive detuning, and vice versa [8]. When the photon energy of the pump light is increased, the negative peak in the vicinity of 3.207 eV decreases. It then disappeared at $\hbar \delta = -5.5$ meV, which suggest the appearance of the upper level. However, due to the complexity of the change in the absorption spectra, the peak energy and its strength could not be identified. Therefore, from the red-shift due to the lower level at the photon energy of 150 meV, we discuss the optical Stark effect.

Figure 5 shows the excitation density dependence of the Stark shift when light with an energy of 150 meV is used. The eigenvalues in the exciton dressed state are given by equations [8] (1) and (2),

$$\hbar \omega_1' = \hbar \left( \omega_1 + \frac{\delta}{2} \right) \pm \hbar \sqrt{\frac{\delta^2}{4} + \Omega_0^2}$$

(1)

$$\hbar \omega_2' = \hbar \left( \omega_2 - \frac{\delta}{2} \right) \pm \hbar \sqrt{\frac{\delta^2}{4} + \Omega_0^2}$$

(2)

where $\delta$ is the detuning from the resonance energy, $\Omega_0$ is the resonant Rabi frequency which is given by $\Omega_0 = |\mu_{12}E_0|/\hbar$ using the electric field intensity $E_0$, the transition matrix element $\mu_{12}$, $\hbar \omega_1$ and $\hbar \omega_2$ are the energies of the two states, respectively, and $\hbar$ is the Dirac constant. By using the Stark shift due to a lower level of obtained from the experiment, the transition dipole moment $|\mu_{12}|$ between 1s and 2p exciton levels is obtained from the equations (1) including the resonant Rabi frequency with $\hbar \omega_1 = 3.202$ eV, $\hbar \delta = 19.5$ meV. Using the result of the excitation density dependence of the Stark shift, the transition dipole moment was obtained for the respective excitation density, and by obtaining the average value, the transition dipole moment $|\mu_{12}|$ was determined as $2.97 \pm 0.09 \text{Å}$. Using the energy shift due to the Stark effect, the transition dipole moment between the 1s and 2p exciton levels in the CuCl single crystal was obtained.
4. Conclusions

The optical Stark effect between the $1s$ and $2p$ exciton levels in CuCl was observed by time-resolved reflection spectrum measurement. A reduction of the reflectance with a time width approximately equal to the pump pulse width was observed from the time-resolved reflection spectrum. Furthermore, from the absorption spectrum obtained from the Kramers-Kronig transform (K-K transform), it was confirmed that the peak energy was red shifted, the absorption amount decreased, and the spectrum width broadened during the time period of pump light irradiation. The transition dipole moment between the $1s$ and $2p$ exciton levels in CuCl were obtained from the excitation density dependence of the Stark shift at the time of pump light irradiation. The future task is to consider the influence of the coupled $2p$ level on the $1s$ level and to study the behavior of the Stark effect in the near resonance region by improving the quality of the sample.

References

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