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Measurement of Photorefractive Two-Beam Coupling Gain of Defect Chalcopyrite Compound $CdGa_2Se_4$

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Photorefractive two beam coupling gain of $CdGa_2Se_4$ was measured as a function of the grating period, the temperature and the pumping intensity. The maximum photorefractive gain of $1.27cm^{-1}$ was obtained at the grating period of 0.446μm. The photorefractive gain exhibited the large temperature dependence which had the maximum at 360K. The response of the amplified signal light to the pumping light pulse was faster than that in GaP.

1. INTRODUCTION

$CdGa_2Se_4$, one of the ternary semi-conducting compounds crystallizing in the defect chalcopyrite structure, has a direct and wide energy gap (2.67eV at 77K). Undoped $CdGa_2Se_4$ crystals grown by the Bridgman method exhibited interesting properties such as the electro-optic effect, the high dark resistivity ($>10^{12}\Omega cm$) and the large photo-conductivity [1]. These features are required for photorefractive material. In our previous paper [2], we reported the first observation of photorefractive effect of $CdGa_2Se_4$.

In this paper, we study the dependence of the photorefractive gain of $CdGa_2Se_4$ on the period of grating, the intensity of excitation light and the temperature by using the two-beam coupling method. The maximum photorefractive gain and the properties related to the photorefractive effect are examined from experimental results. The response time of the photorefractive effect is also measured.

2.OPTICAL ARRANGEMENT OF TWO BEAM COUPLING METHOD

$CdGa_2Se_4$ crystals were oriented and cut to plates with the (001) faces. A (001) plate was cut into rectangular prisms with the faces perpendicular to two new axes induced by an electric field applied along the [001] axis as shown in Fig.1. This configuration is similar to that used for the photorefractive measurement of zincblende semiconductors because the non-diagonal component γ_{14} of the electro-optic coefficient is ignored as compared to γ_{63}. Both signal beam and pump beam have the s-polarization for the interference in the crystal and the grating space-charge field occurs along the [001] axis.

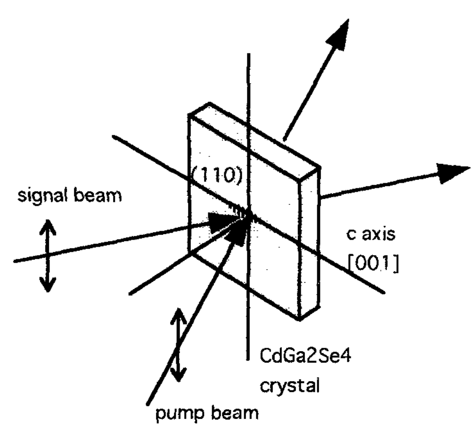

Fig.1 Configuration of $CdGa_2Se_4$ crystal and light beams for two beam coupling experiment.

The experimental setup for measuring the gain coefficient Γ is shown in Fig.2. A signal beam and a pump beam of a He-Ne laser (633nm,45mW) are divided by a beam splitter and are interfered in the $CdGa_2Se_4$ crystal (1.2mm thickness) with (110) face. The gain coefficient Γ is determined by the following equation on the assumption that the signal beam intensity is much smaller than the pump intensity,

$$\Gamma = \frac{\cos\theta}{L}\ln\left(\frac{I_s + \Delta I_s}{I_s}\right), \qquad (1)$$

where Is is the intensity of the signal beam in the absence of pump beam and ΔIs is the amplified intensity of the signal beam through the interaction with the pump beam. θ is the half angle between the pump beam and the signal beam in the crystal, and L is the thickness of the crystal. In a conventional optical setup for two-beam coupling measurement, a chopper is inserted into the pump beam and the change of the signal beam only caused by the pump beam is detected by a photomultiplier synchronized with the chopper. However, the absorption coefficient largely depends on the temperature in the $CdGa_2Se_4$ crystal. The intensity of signal beam can be changed by the temperature rise due to irradiation of the pump beam. To measure only the photorefractive effect by suppressing other effects, we adopted a phase modulator in stead of a chopper. It produces alternately the s-polairzed and the p-polarized pump beam. As the signal beam is s-polarized, the s-polarized pump beam can induce the photorefractive grating as a result of the interference but the p-polarized pump beam cannot. As the intensity of pump light is not changed by the phase modulation, the change of the absorption coefficient by the irradiation of the pump beam is constant, and there, it is not amplified by the lock-in amplifier synchronized to the phase modulator.

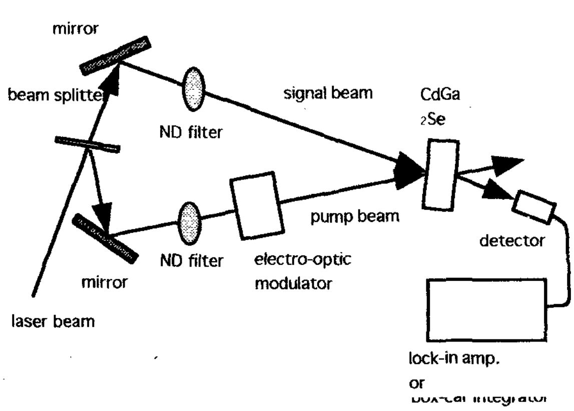

Fig.2 Experimental setup for two-beam coupling measurement.

3.PHOTOREFRACTIVE GAIN Γ OF $CdGa_2Se_4$

3.1 Grating wave number

The photorefractive gain Γ was measured at the different angles of θ by using the experimental setup in Fig.2 at room temperature. Figure 3 shows the experimental results of Γ as a function of the grating wavenumber k ($=2\pi n\cos\theta/\lambda$, n:refractive index of the sample). Γ takes the maximum value of $1.27cm^{-1}$ at k of $14.08\mu m^{-1}$ ($\Lambda=0.446\mu m$) at 300K.

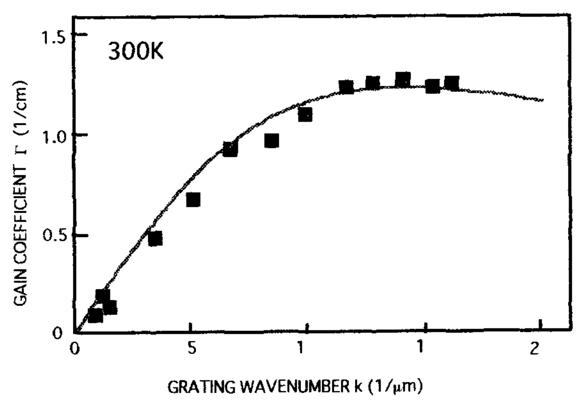

Fig.3 Photorefractive gain coefficient Γ as a function of the grating wavenumber k.

The photorefractive gain Γ of the crystal without any external electric field is expressed by

$$\Gamma = \frac{2\pi n^3 \gamma_{eff}}{\lambda\cos\theta}\frac{E_d E_q}{E_d + E_q}, \qquad (2)$$

where

$$E_d = \frac{k_B T}{e} k \quad \text{and} \quad E_q = \frac{e N_E}{\varepsilon} \frac{1}{k} , \qquad (3)$$

E_d is the diffusion field, E_q is the impurity density-limited space charge field and N_E is the effective trap density. Γ takes the maximum value in general case when $E_d=E_q$. Then, the grating period Λ(=2π/k) is given from eq.(2) by

$$\Lambda_{max} = 2\pi \sqrt{\frac{\varepsilon k_B T}{e^2 N_E}} \quad . \qquad (4)$$

The effective trap density N_E is found to be $2.3x10^{15}cm^{-3}$ from the experimental result of Λ at the maximum Γ using eq.(4). From the value of N_E and the refractive index n (=2.7), Γ is calculated as a function of wave numbers k (=2πncosθ/λ) using eq.(2). It agrees with the measured values in case of $\gamma_{eff}=3.5x10^{-12}m/V$ as shown in Fig.3 by the solid curve.

3.2 Temperature

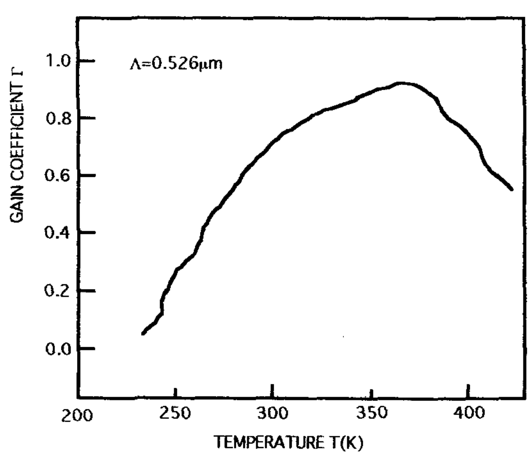

Fig.4 Temperature dependence of the gain coefficient Γ.

The photorefractive gain was measured at various temperatures on the constant grating period (Λ=0.526μm). Figure 4 shows the experimental results of the dependence of the gain coefficient Γ on temperature. Γ increases from 230K and takes the maximum value at 360K. It decreases with temperature above 360K. The decrease in the high tem-perature region which has been generally observed on other semiconducting materials is thought to be due to screening by thermally excited carriers. We already reported the thermal quench of photocarriers of $CdGa_2Se_4$ in the temperature region from 190K to 350K[1]. It is attributed to the recombination between electrons in the conduction band and holes thermally released to the valence band from hole trap centers. The decrease of Γ in low temperature region is thought to be due to the same mechanism of the thermal quench of photocurrent.

3.3 Pumping intensity

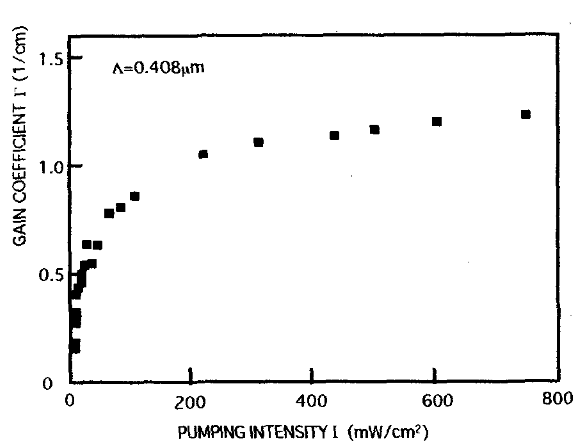

Fig.5 Dependence of photorefractive gain Γ on pumping intensity I

The photorefractive effect is characterized from other nonlinear optical effects by the property that the gain coefficient is independent of the intensity of pump beam. However, it depends practically on the pumping intensity when the pumping light is weak. The saturation property of Γ is related to the transport and the storage mechanism of photogenerated carriers. Figure 5 shows the gain coefficient Γ measured on the different excitation intensities. The saturation property of Γ is defined by

$$\Gamma = \frac{\Gamma_\infty}{1+(I_s/I)} \quad , \tag{5}$$

where Γ is the saturated gain and I_s is the saturated pumping intensity. As a result of fitting of eq.(5) to experimental results, it was found that the gain coefficient, Γ=1.3cm^{-1} ,and Is took the two kinds of values of 15 and 43mW/cm^2. This result means that photorefractive effect of $CdGa_2Se_4$ is caused by two mechanisms of the storage of photogenerated carriers.

4. RESPONSE OF PHOTOREFRACTIVE EFFECT

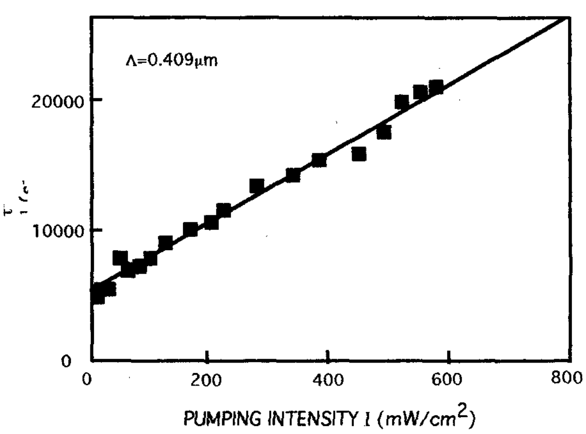

Fig.6 Response time of photorefractive effect as a function of the intensity of pump beam

The waveform of the amplified signal beam is measured by using a boxcar integrator when the electric field pulse was applied to the phase modulator. The response time was determined from the decay of the signal beam.

As a result of measurement on the various pumping intensities as shown in Fig.6, the following relationship between the response time (s) and the pumping intensity I (mW/cm^2) was obtained.

$$\tau^{-1} = 27 \cdot I + 5200 \tag{6}$$

The experimental result shows that $CdGa_2Se_4$ has much faster response time of the photorefractive effect than GaP[3].

5. CONCLUSION

The photorefractive effect of $CdGa_2Se_4$ was studied by using the two-beam coupling method. The following results were obtained: The photorefractive gain take the maximum value of 1.27cm^{-1} at the grating period of 0.446μm at 300K. It has the large temperature dependence which exhibits the maximum at 360K. The measured response time of photorefractive effect is much faster than that of GaP. These results show that $CdGa_2Se_4$ is a potential semiconducting material for applications of photorefractive effect. However, the optical quality of our $CdGa_2Se_4$ crystals is not sufficient for practical applications at present. The crystal growth conditions have to be investigated further to obtain $CdGa_2Se_4$ crystals without absorption loss.

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