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Absorptive optical bistability due to impurity nonlinearity in CdS : Cu

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With the excitation of a ps pulse laser we found both the optical bistability and the multi-order bistability in CdS : Cu^+ crystals. The nonlinear response time is as fast as 150 ps, and the switching time of the second-order bistable loop approaches 80 ps. The technique of the degenerate four wave mixing (DFWM) has been used to investigate the optical nonlinearity related to the Cu^+ impurity. The third-order nonlinear susceptibility of $\chi^{(3)}$ was measured to be 9×10^{-9} esu.

1. Introduction

Various optical nonlinearities have been observed in undoped CdS, mostly due to intrinsic effects such as exciton–exciton scattering, biexciton creation, and the occurrence of an electron–hole plasma [1]. However, the impurity nonlinearities play a key role in the bound-exciton energy range for realizing the optical switching processes and optical bistability [2–4].

The transmission behaviour of In-doped CdS in the spectral range below the band gap was reported by Hönig et al. [5]. They pointed out that indium formed shallow acceptor and donor bands at a high impurity concentration. The investigated CdS : In crystals with In concentration of 5×10^{18} cm^{-3} exhibited a strong broad band absorption due to the transition from the acceptor into the conduction band. The switching times decreased with increasing excitation density and laser wavelength. Strauss et al. [6] reported the photorefractive investigation of the dynamics and relaxation of photoexcited carriers in semiconductor CdS doped with Cu^+. The Cu^+ centers introduced the deep energy level inside the bandgap.

The present paper will report on the bistability and the multi-order bistability relevant to Cu^+ impurity in Cu-doped CdS. The method of the degenerate four wave mixing (DFWM) is employed to study the impurity nonlinearity. It is found that the third-order nonlinear susceptibility of $\chi^{(3)}$ is increased by 3 orders of magnitude compared with undoped CdS. This is attributed to the transition from the Cu^+ level to the conduction band. For high excitation we find the optical bistability and the two-order bistability, and for the second-order hysteresis loop the switching time is obtained to be as short as 80 ps.

2. Basic theory on $\chi^{(3)}$

The theoritical expression of the third-order nonlinear susceptibility of $\chi^{(3)}$ will begin with Maxwell's equations. The common diagram of the phase conjugation for DFWM is shown in fig. 1.

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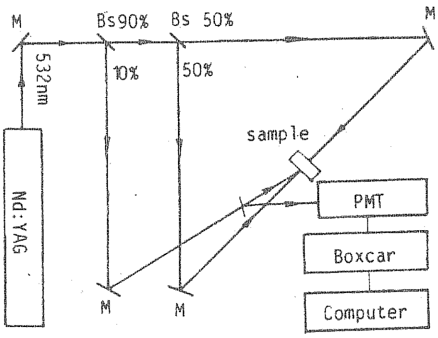

Fig. 1. Experimental configuration for DFWM Bs: beam splitter; M: reflection mirror; PMT: photomultiplier.

In the case of the steady state using the slow-varying envelope approximation, the coupled equations are given by

$$\frac{\mathrm{d}E_3^*}{\mathrm{d}z} = -\mathrm{i}\beta_0\,\mathrm{e}^{-\alpha L/2}E_4 - \frac{\alpha}{2}E_3^*,$$

$$\frac{\mathrm{d}E_4}{\mathrm{d}z} = \frac{\alpha}{2}E_4 - \mathrm{i}\beta_0^*\,\mathrm{e}^{-\alpha L/2}E_3^*,$$

having supposed

$$E_1(z) = E_{10}\,\mathrm{e}^{-\alpha z/2},$$

$$E_2(z) = E_{20}\,\mathrm{e}^{\alpha(Z-L)/2},$$

combined with the boundary conditions of $E_3(0) = E_0$ and $E_4(L) = 0$, where L denotes the sample thickness. Then,

$$E_3(z) = \frac{2E_0\left[-\frac{\alpha}{2}\sin\beta(Z-L) + \beta\cos\beta(Z-L)\right]}{\alpha\sin\beta L + 2\beta\cos\beta L},$$

$$E_4(z) = \frac{-2\mathrm{i}E_0\beta^*\sin\beta(Z-L)\,\mathrm{e}^{-\alpha L/2}}{\alpha\sin\beta L + 2\beta\cos\beta L}.$$

The reflection of phase conjugation is defined as $R = |E_4(0)|^2/|E_3(0)|^2$. If considering a small signal intensity, then

$$\chi^{(3)} = \frac{2n^2c^2\epsilon_0}{3\omega l}\left(\frac{R}{I_1 I_2}\right)^{1/2},$$

where $l = (2L\,\mathrm{e}^{-\alpha L/2})/(\alpha L + 2)$ is called the effective interaction length; I_1 and I_2 are the forward and backward pump intensity.

3. Experimental

For the nonlinear measurement, the configuration of DFWM is shown in fig. 1. A 532 nm line from a Q-switched Nd : YAG laser is split into three beams. A weak probe beam and two equally intense pump beams with the counterpropagation direction are incident on the sample simultaneously. The probe beam is incident at a small angle of 3° to 5° with respect to the forward pump light. The signal of DFWM is detected by a photomultiplier.

For the bistable experiment, a mode-locked Nd : YAG is used as an excitation source. The pulse duration varies from 150 to 1000 ps. One beam transmitting the crystal is taken as the pump light, and the other as reference. The two beams are received by a streak camera with a time resolution of 2 ps. They are analysed and processed by a personal computer.

The investigated crystal CdS is doped with Cu^+ in the concentration of 10^{19} cm^{-3} during the crystal growth process.

4. Results and discussion

The absorption and luminescence spectra of CdS : Cu at room temperature are shown in fig. 2

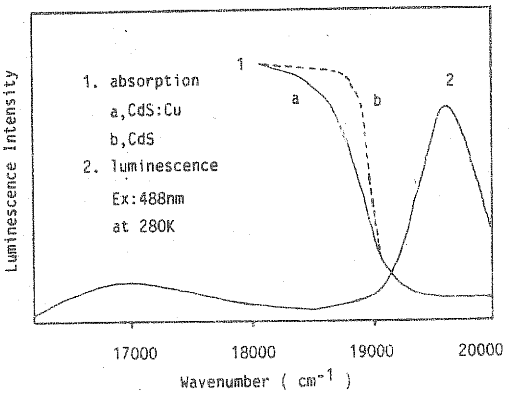

Fig. 2. Spectra of absorption and luminescence in CdS : Cu.

with an excitation of 488 nm. In the visible range of the spectrum, one finds two emission bands which originate from the band edge emission of CdS and the transition from the conduction band to the Cu^+ deep level respectively. In the present concentration of Cu^+ the main excitation process at 532 nm is to give rise to the ionization of Cu^+ centers. Because of considerable contents of other impurities and native defects in CdS, they may make some contribution to the absorption, and critically affect the kinetic processes. Therefore, CdS : Cu will exhibit the strong nonlinearity related to impurities. Figure 3 shows the dependence of the phase conjugation reflectivity of R on pump intensity of DFWM. $\chi^{(3)}$ is measured to be 9×10^{-9} esu for CdS : Cu and 3×10^{-12} esu for undoped CdS at 532 nm. The nonlinear susceptibility due to impurity existence is increased by three orders of magnitude with respect to the undoped case.

The transmission behaviour in a variety of excitation densities with 532 nm excitation is presented in fig. 4. The incident pulse with the duration of 300 ps is compressed to be 200 ps as shown in fig. 4(a). If the transmitted intensity is drawn as a function of the incident intensity, a counterclockwise hysteresis loops can be obtained as shown in fig. 4(b). The switching times are measured to be $t_\uparrow = 150$ ps and $t_\downarrow = 200$ ps.

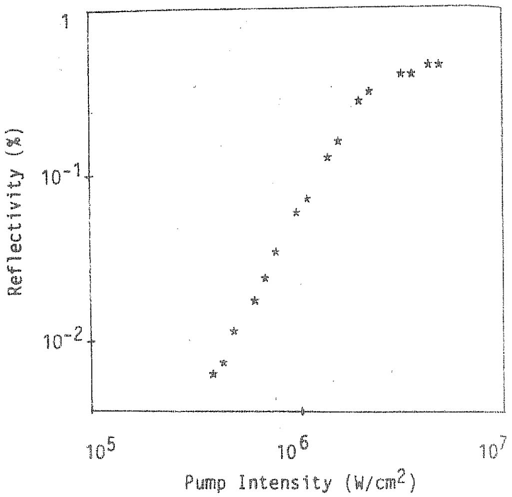

Fig. 3. Relationship of conjugation reflection versus pump intensity.

Furthermore, we find the two-order bistability while the excitation density is suitable. The second bistable cycle shows the fast switching time to be 80 ps.

The optical bistability can be understood with the absorption between the Cu^+ level and the conduction band, while the positive feedback is provided by the light reflection on the crystal surfaces. In the present spectral range the reflec-

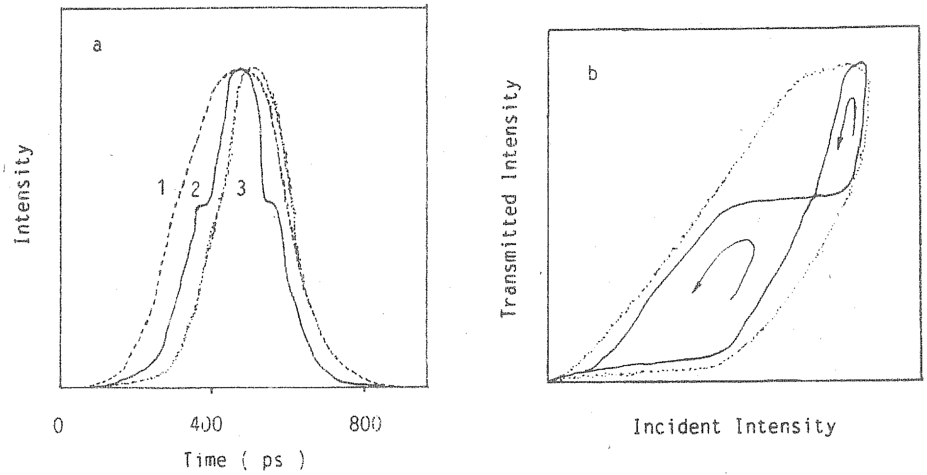

Fig. 4. Comparison of pulse shapes of the transmitted and incident laser for pulse duration of 300 ps. 1: incident pulse, 2 and 3: transmitted pulses; excitation density of 7 MW/cm^2 for case 2 and 15 MW/cm^2 for case 3.

tion from perfect surfaces is over 30%. According to Henneberger's calculation [7], this value is sufficient to obtain the bistability considering the strong impurity absorption. The multibistability may be interpreted by the character of the Fabry–Perot cavity. Using the graphical solution [8], one knows the transmission as a function of phase B

$$T = I_t/I_i = 1/(1 + F \sin^2(B/2)).$$

Then, the transmission can also be written as

$$T = I_t/I_i = B_1 I_t/(B - B_0).$$

B is given by $B = B_0 + B_1 I_i$. The solutions of I_t and I_i must satisfy both relations mentioned above. Hence, the multibistability can be established when the incident intensity is high enough.

References

[1] I. Broser and J. Gutowski, Appl. Phys. B 46 (1988) 1.
[2] T. Hönig, J. Gutowski and I. Broser, J. Cryst. Growth 86 (1988) 576.
[3] T. Hönig and J. Gutowski, Phys. Stat. Sol. (b) 150 (1988) 833.
[4] G.O. Müller, S.L. Karpenko, R.P. Seisyan and M.A. Yakobson, Phys. Stat. Sol. (b) 120 (1988) K93.
[5] T. Hönig, J. Gutowski and I. Broser, J. Lumin. 45 (1990) 194.
[6] E. Strauss, Th. Sommer and R.K. Germer, Proc. Photothermal Phenomena Meeting '89, in Baltimore (Springer, Berlin, 1990).
[7] F. Henneberger, Phys. Stat. Sol. (b) 137 (1986) 371.
[8] H.M. Gibbs, Optical Bistability: Controlling Light with Light, eds. P.F. Liao and P. Kelley (Academic Press, New York, 1985).