Absorptive optical nonlinearity and bistability in semimagnetic seminconductor $Zn_{1-x}Mn_xSe$

Dou Kai, Huang Shihua, Qin Weiping, Jin Chunming, Zhou Fangce, Yu Jiaqi, Zhang Xinyi, and Xu Xurong

窦 恺 黄世华 秦伟平 靳春明 周方策 虞家琪 张新夷 徐叙瑢

Changchun Institute of Physics, Academia Sinica, Changchun, 130021

(Received 15 June 1990; revised 28 April 1991)
Chin J. Semicond. **12**(12), 759–762 (December 1991)

Semimagnetic semiconductor $Zn_{1-x}Mn_xSe$ ($x = 0.01$) exhibits a broad-band absorption in the region of the band edge. The absorption is easily blue-shifted with the pump-probe system. Optical bistability is obtained for the first time in a wider range of excitation wavelengths. The origins of the nonlinear and bistable effects are discussed.

I. INTRODUCTION

The highly interesting optical and magneto-optical properties of semimagnetic semiconducting materials (Zn, Mn) Se have attracted the attention of many research scientists and these semiconductors have undergone widespread investigations and thorough studies.[1,2] Among these kinds of semiconductor materials, the Group II elements in the II–VI compounds form solid solutions in which they are replaced by the magnetic ion Mn^{2+}. The widths of the energy gaps for (Zn, Mn) Se compounds depend on the compositional values of the Mn ion, Mn^{2+}, or on the externally tunable magnetic field in the case of a fixed Mn composition. We have not only studied the absorption and the emission properties of the ternary semiconductor compound $Zn_{1-x}Mn_xSe$, but also investigated thoroughly its optical nonlinear characteristics.[3] This paper will report for the first time the experimental results on the nonlinear optical absorption and the bistability phenomenon in $Zn_{1-x}Mn_xSe$. The unique nonlinear optical properties of $Zn_{1-x}Mn_xSe$ make it useful in many applications such as optical signal processing, optical computing, and optical storage, etc.

II. EXPERIMENTAL METHOD

The $Zn_{1-x}Mn_xSe$ single-crystal samples used in the present experiment were prepared by the vapor sublimation process. The Mn fraction x was measured to be $x = 0.01$ by the x-ray diffraction method. The single crystal was cleaved into thin wafers with thicknesses varying from 0.5 mm to 0.9 mm according to crystal cleavage plane. When the two surfaces of the wafers were parallel, combined reflectivities of about 80% could be obtained. The absorption spectra were measured at different temperatures using a closed-cycle liquid-helium cryogenic cooler, the temperature range of which was controlled from 10 K to 300 K.

The nonlinear optical absorption measurements were performed using a pump/probe system. The laser source used for studying the bistability experiment was a pulsed Nd:YAG laser and a pump dye laser with a wavelength of 532–565 nm. The laser pulse width was smaller than 20 ns; the laser intensity was highly stable and the pulse-to-pulse repeatability was excellent. After the beam was transmitted through the sample and the incident optical pulse was detected by a high-speed photodiode, it was finally displayed on an oscilloscope screen. The time resolution of the entire measurement system could reach 500 ps.

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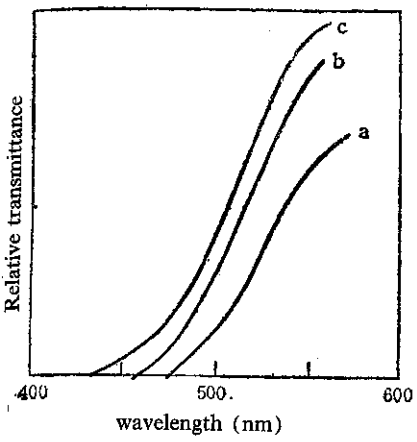

FIG. 1. The transmission curves for different pump intensity levels. (a) Zero pump intensity; (b) pump intensity $= 8\times10^4$ W/cm^2; and (c) pump intensity $= 4\times10^5$ W/cm^2.

III. RESULTS AND DISCUSSIONS

1. Nonlinear optical absorption

The nonlinear optical absorption was observed using a pump wavelength ranging from 532 nm to 560 nm. For different pump intensities, the transmittance of the sample was measured. The results are shown in Fig. 1; the pump-laser wavelength was set at 532 nm. In the figure Curve (a) denotes the relative optical transmission through the sample with no pumping. Curves (b) and (c) denote the relative transmissions for pump intensities of 8×10^4 W/cm^2 and 4×10^5 W/cm^2, respectively. It can be seen clearly that as the pump intensity is raised, the transmission through the sample rises and the absoprtion edge apparently shifts toward the higher energy side.

The absorption spectral characteristics of $Zn_{1-x}Mn_xSe$ differ from those of ZnSe; the Mn composition in the semiconductor determines the energy band gap.[4] The authors in Ref. 4 studied the optical properties of $Zn_{1-x}Mn_xSe$ with a relatively high fractional content of Mn, $x\geqslant0.1$. These authors pointed out that as the content of Mn increased the energy band gap became wider. However, in our own study on $Zn_{1-x}Mn_xSe$ crystals with low values of x such as $x = 0.01$, we were surprised to find that the observed energy band gap was actually smaller than that of ZnSe. We have not been able to provide a reason for this discrepancy up to the publication submission date of this paper. At room temperature, $Zn_{1-x}Mn_xSe$ crystals exhibit band edge absorptions such as those shown in Fig. 2(a). When the temperature is reduced to 200 K, the absorption edge apparently shifts towards the high-energy direction; this is accompanied by an absorption band near 19,000 cm^{-1} or 526 nm. As the temperature is reduced further, an absorption band near 20,000 cm^{-1} or 500 nm then appears. The positions of these two absorption bands do not change with temperature [see Fig. 2(b)]. Based on the discussions given in Ref. 5, they are related to the $^6A_1\rightarrow{}^4T_1$ and $^6A_1\rightarrow{}^4T_2$ transitions in Mn^{2+}. When the 532 nm laser wavelength is used for the excitation of the sample, the absorption originates at the band edge and the $^6A_1\rightarrow{}^4T_1$ electronic transition. Under weak excitation conditions, this transition leads to the relatively strong absorptuion in $Zn_{1-x}Mn_xSe$. As the laser excitation power increases, many more electrons are excited to the upper energy levels, hence the appearance of the "blue" shift.

For a fixed laser wavelength at 532 nm and various laser power excitations, the transmissions and incident wave shapes and their dependence on pump laser intensity are shown in Figs. 3(a) and 3(b), respectively. Figure 3(a) shows the measurement results for the pulse shapes of the transmitted optical beam intensity (I_T) and the incident optical beam intensity (I_0). The transmitted laser pulse clearly shows a definite distortion and a time delay. As the excitation intensity increases, the pulse-shape delay increases further. If the transmitted laser beam intensi-

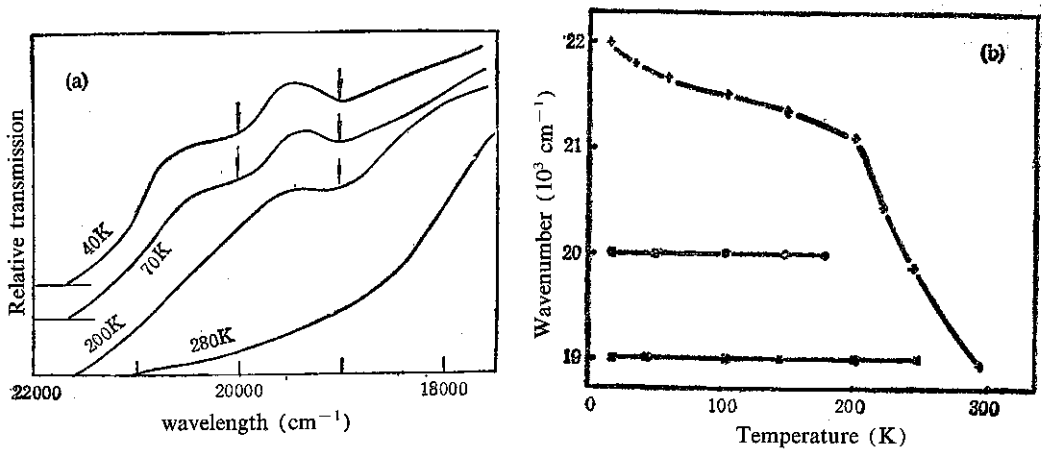

FIG. 2. Absorption spectra taken at different temperatures. (a) The arrows enote the locations of 500 nm and 526 nm absorptions related to the Mn^{2+} energy level absorption bands. Vert. axis label = relative transmittance; (b) The 500 nm 526 nm, and band-edge absorptions as functions of temperature: pulses = absorption edge; solid circles = 500 nm; crosses = 526 nm.

ty is assumed to be a function of the incident laser beam intensity and the result is plotted, we obtain the curves shown in Fig. 3(b) showing the bistability loop with the direction changing with time. When the excitation power was 10^6 W/cm^2, we obtained the bistability switching times of $\tau_\uparrow = 6$ ns and $\tau_\downarrow = 800$ ps. When the excitation wavelength was 560 nm and the excitation intensity was 10^5 W/cm^2, similar measurements resulted in switching times $\tau_\uparrow = 9$ ns and $\tau_\downarrow = 1.8$ ns. Recently, we have used the "picosecond laser and streak camera" modern spectroscopic technique and determined through measurement that the bistability switching time was about 150 ps.[6]

The origin of the bistability phenomenon in optical absorption in $Zn_{1-x}Mn_xSe$ crystals stems from the relatively strong nonlinear positive feedback provided by the optical reflection process occurring in both the bulk crystal and at the crystal surface of the semiconductor. The bistability process can be described as follows: when the incident optical intensity is relatively weak, the sample exhibits linear absorption and the system is best described as being situated in a high absoprtion state; as the incident optical intensity is raised, the transmission through the sample gradually increases, and the absorption decreases relatively, and the system is situated in a high-transmission state; the high-transmission state remains intact through the interval in which the incident optical intensity changes from strong to weak, and the positive optical feedback supplied by the crystal surface of the sample keeps the system in the high-transmission state; finally, when the incident optical intensity decreases further and the absorption gradually

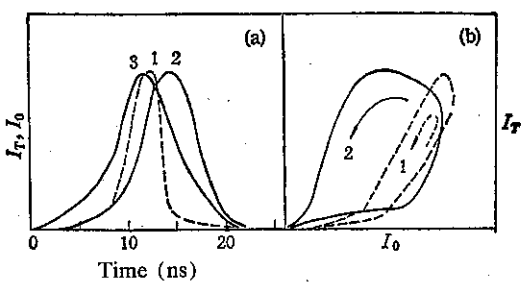

FIG. 3. The transmission and incident pulse shapes and their bistability. (a) Comparison of the transmitted and incident pulse shapes; (b) the transmitted beam intensity as a function of the incident beam intensity.

increases, the positive feedback can no longer maintain the high transmission state; this allows the system to return to the high absorption state from the high transmission state.

IV. CONCLUSIONS

$Zn_{1-x}Mn_xSe$ crystals exhibit strong nonlinear optical absorption and optical bistability phenomena over a rather large range of excitation wavelengths. The energy transitions in Mn^{2+} lead to the nonlinear absorption. The bistability phenomenon originates from the material nonlinearity, and the surface reflection supplies the positive feedback. Both the excitation power density and the excitation wavelength play important roles in the bistability phenomenon.

The authors would like to thank colleagues Xuan Li, Ling Zhihua, Jing Yumei, and Huang Ximin for supplying the crystal samples, and helpful discussions.

[1]J. K. Furdyna, J. Appl. Phys., **53**, 7637 (1982).
[2]D. H. Auston *et al.*, Applied Optics **26**, 211 (1987).
[3]Dou Kai, Huang Shihua *et al.*, Solid State Commun. **76**, 1165 (2990).
[4]R. B. Bylsma, W. M. Becker, J. Kossut, U. Debaka, and D. Yoder-Short, Phys. Rev., **B33**, 8207 (1986).
[5]D. Langer and H. Richter, Phys. Rev., **146**, 554 (1966).
[6]Dou Kai, Huang Shihua *et al.*, to be published.

Translated by A. K. Eng
Edited by G. M. Temmer