



Mn-RELATED ABSORPTIVE OPTICAL NONLINEARITY AND BISTABILITY IN $\text{Zn}_{1-x}\text{Mn}_x\text{Se}$

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ZnMnSe exhibits a strong nonlinear absorption related to Manganese ions. This absorption is easily bleached when the another narrow-band laser with high intensity illuminates the crystals. The first observation of the absorptive optical bistability is observed in a wider range of excitation power density and excitation wavelength. It is thought that the bistable effect is attributed to the bleaching of absorption concerning Mn energy bands combined with the positive-feedback provided by the light reflection at the perfect crystal surfaces.

INTRODUCTION

The interesting optical magneto-optical properties of monocrystals $(\text{Zn,Mn})\text{Se}$ have been extensively studied by many authors[1,2]. These materials are II-VI binary compounds, in which a magnetic ion is substitutionally incorporated in the host crystals in place of group-II element. $\text{Zn}_{1-x}\text{Mn}_x\text{Se}$ is a wide-band-gap semimagnetic semiconductor whose band gap is tunable compositionally and magnetically. The structure of energy band and crystal of this kind of alloy system depends on Mn component.

Recent advances in semiconductor growth techniques have made it possible to grow semiconductor structure with atomic-scale control of the composition and thickness[3]. For the quantum-well structures, the position of the electronic energy levels is modulated, and the carriers are confined in the lowest band-gap medium. The quantum-size effects significantly modify the electronic and optical properties of these materials compared with those of the bulk materials. But so far, no reports on the bistable effects in bulk crystals $(\text{Zn,Mn})\text{Se}$ is given.

Currently we report the first work on the nonlinear optical property and the bistable phenomenon of the bulk monocrystal $\text{Zn}_{1-x}\text{Mn}_x\text{Se}$ with considerably low Mn composition ($x=0.01$). The absorptive optical nonlinearity and bistability are carefully investigated in a wide range of excitation wavelength and power density, and this bistable is attributed to the nonlinear absorption concerning Mn energy band combined with the positive-feedback provided by the light reflection from the crystal surfaces. In view of our results, all of these features are very promising for application to optical signal generation, processing, and transmission.

EXPERIMENTAL

The investigated crystals $\text{Zn}_{1-x}\text{Mn}_x\text{Se}$ are

platelets with thickness of 0.5-0.9mm, and with a small composition of x determined to be 0.01. The crystals are prepared by the vapor sublimation, and separated into pieces of platelets in the direction of crystal planes.

Sometimes, samples are placed in the cryogenic refrigeration system in which the temperature is available to regulate continuously within the range of 10-300K. The pump-probe system is employed to study the nonlinear absorption. The high excitation density is changed by using a set of neutral filters.

The bistable experiment are carried out with a 532nm line of a Nd:YAG laser or a dye laser with a tunable wavelength range of 550-565nm. The duration of laser pulses is not more than 7ns. The laser pulses transmits the crystals, and the transmitted signals and the incident laser are simultaneously detected by a fast photocell or photodiode. Both of them are displayed on the screen of an oscilloscope with the time resolution of 500ps.

RESULTS AND DISCUSSION

1. Absorptive optical nonlinearity

The Manganese-related absorptive nonlinearity is observed in an excitation wavelength range from 532 to 560nm. The nonlinear absorption as a function of pump laser is shown in Fig.1 for the excitation wavelength of 532nm. The curve (a) stands for the unpumped case, and the curve (b) and (c) represent the situation with high excitation density of 8×10^4 and $4 \times 10^5 \text{ W/cm}^2$, respectively. It is clearly seen that with increasing excitation intensity, the crystals exhibit an increasing transmission, and the absorption edge shifts to the higher energy, i.e., a blue shift, within the whole spectral range.

Spectral properties of ZnMnSe are not similar to ones of ZnSe , and characterized by

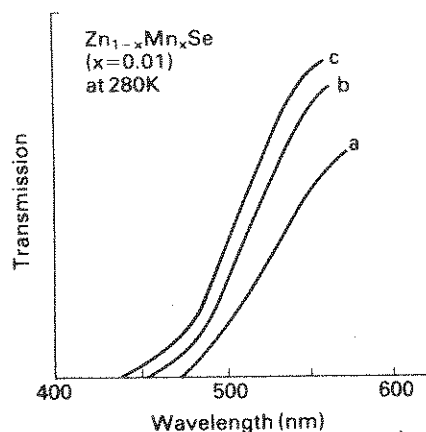


Fig.1. Absorption spectra at a variety of excitation densities with a 532nm pumping at 280K. a, unpumped case; b, $8 \times 10^4 \text{ W/cm}^2$; c, $4 \times 10^5 \text{ W/cm}^2$.

a broad-band absorption below the energy-band-gap of ZnSe at higher temperature, and featured by two absorption bands at lower temperature as shown in Fig.2. Two bands indicated by arrows are observed at 40K and 70K, and center at 20000 cm^{-1} (500nm) and 19000 cm^{-1} (526nm). The position of the absorption bands do not depend on temperature. We attribute the two bands to the transition of ${}^6\text{A}_1 \rightarrow {}^4\text{T}_1$ and ${}^6\text{A}_1 \rightarrow {}^4\text{T}_1$ of Mn^{2+} ion[4].

The bleaching of the absorption can be interpreted by the transition from ${}^6\text{A}_1$ to ${}^4\text{T}_1$ (526nm), just mixed in the absorption edge at room temperature in the unexcited case. Under low excitation intensity, the transition between ${}^6\text{A}_1$ and ${}^4\text{T}_1$ results in a strong absorption. As the excitation grows, much more electrons fill in the top energy level. Hence, a bleaching of absorption can be observed.

2. Absorptive optical bistability

Fig.3 shows the absorptive bistability for a laser of 532nm at different excitation densities up to 1 MW/cm^2 . The pulse shapes of the transmitted and the incident laser pulses are given in Fig.3a. The transmitted pulse shapes show the deformity and delay compared to the reference laser pulse. If the transmitted intensity is drawn as a function of the incident intensity, a counterclockwise hysteresis loop results from the waveform delaying as shown in Fig.3b. We measure the switching times $\tau_1 = 6 \text{ ns}$ and $\tau_2 = 800 \text{ ps}$ at 1 MW/cm^2 . And also, the bistable phenomenon can be obtained in a pump wavelength region from 532 to 560nm. A similar measurement for 560nm excitation gives the switching time to be $\tau_1 = 9 \text{ ns}$ and $\tau_2 = 1.8 \text{ ns}$ at 10^5 W/cm^2 . Furthermore, it is found that a decrease of the excitation wavelength results in a decrease of the switching energy, and the switching time drops with increase of excitation power density.

The absorptive bistability can be understood in the model of bleaching of absorption,

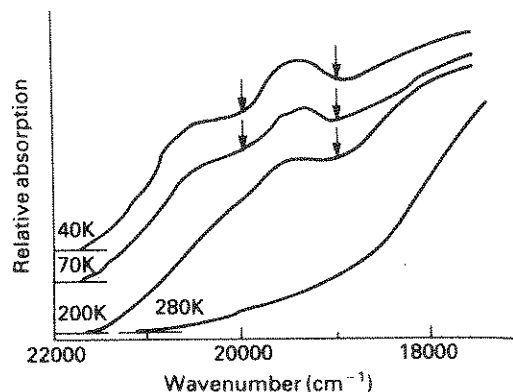


Fig.2. Temperature dependence of absorption spectra. The arrows indicate two bands of absorption from Mn ions.

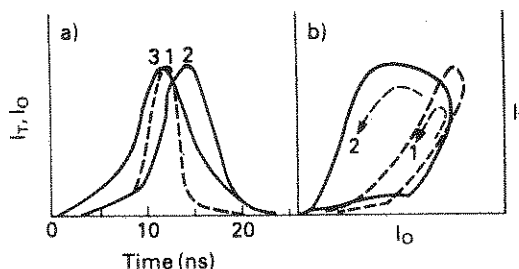


Fig.3. Pulse shapes and hysteresis loops at various excitation densities for pump wavelength of 532nm at 280K. 1, 0.5 MW/cm^2 ; 2, 1 MW/cm^2 ; 3, incident reference laser.

while the positive-feedback is provided by the light reflection from the perfect crystal surfaces. The bistable process is explained as follows. The nonlinear absorption and the positive feedback are able to maintain the bistability. Before and even after the incident laser intensity reaches a maximum, the sample remains in a state of high absorption. With decreasing of input intensity and increasing absorption, the positive feedback can not continue holding the stable state. As a result, the system switches back to the lower transmission state from the high transmission.

CONCLUSIONS

In summary, ZnMnSe is a kind of very promising materials for application to the optical bistable device. The bleaching of absorption is associated with the Mn ions. The bistable effect comes from the nonlinear absorption combined with the positive feedback. The excitation wavelength and power density have a notable impact on the nonlinearity and bistability.

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