Proc. Int. Conf. Optical Properties of Nanostructures, Sendai, 1994
Jpn. J. Appl. Phys. Vol. 34 (1995) Suppl. 34-1, pp. 170-172

Measurement of Nonlinear Refractive Index and Nonlinear Absorption Coefficient Using Transmission Spectrum

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We present a simple, but highly sensitive, method for obtaining both the sign and magnitude of the nonlinear refractive index and the nonlinear absorption coefficient as a function of wavelength. It is shown theoretically that these parameters can be easily obtained from the linear and nonlinear transmission spectra of the medium as well as the laser intensity distribution on the frequency. This technique is demonstrated for ZnSe-ZnS multiple quantum wells on transparent CaF_2 substrate at 77 K.

KEYWORDS: nonlinear refractive index, nonlinear absorption coefficient, transmission spectrum

1. Introduction

Being of large refractive nonlinearities associated with excitonic effects at room temperature, semiconducting superlattices and quantum-well structures have attracted much interest recently.[1] They have been promising candidates for producing optical bistable switches, semiconductor diode lasers, optical information processing and other nonlinear optical devices with low-power, high speed and small size. The nonlinear refractive index is a very important parameter in designing such devices.[2] However, there is not yet a satisfactory method to measure the nonlinear refractive index for thin film materials.

Previous measurements of the nonlinear refraction used a variety of techniques including nonlinear interferometry,[3] degenerate four-wave mixing,[4] nearly degenerate three-wave mixing,[5] pump-probe,[6] ellipse rotation,[7] beam distortion measurements,[8] and Z-scan.[9] The first four methods are potentially sensitive techniques, but they require relatively complex experimental apparatus. Beam distortion measurements, on the other hand, are relatively insensitive and needs detailed wave propagation analysis. The Z-scan technique require that the laser be a Gaussian TEM_{00} beam. All these methods are difficult to be used for measuring the nonlinear refractive index in a large wavelength range, especially for thin film samples.

In this paper we propose a new method to determine the nonlinear refraction and nonlinear absorption from transmission spectra. This method is suited especially to semiconductor quantum wells structures, and offers simplicity as well as very high sensitivity. Our method is based on the idea that the nonlinear absorption being a function of wavelength can be obtained from linear and nonlinear transmission spectra, and the nonlinear refraction can then be calculated from the nonlinear absorption using the Kramers-Kronig relations.

2. Theory

For the intensity dependent third order nonlinear absorption process in the small signal regime, the absorption coefficient can be expressed as

$$\alpha(\lambda)=\alpha_0(\lambda)+\triangle\alpha(\lambda)=\alpha_0(\lambda)+\beta(\lambda)I(\lambda) \quad (1)$$

where $\alpha_0(\lambda)$ and $\beta(\lambda)$ are the linear and nonlinear absorption coefficients, respectively, $I(\lambda)$ is the intensity of the laser beam within the sample, and λ is the wavelength of the laser.

The linear transmissive intensity is given by

$$I_L(\lambda)=\frac{1}{C}I(\lambda)exp[-\alpha_0(\lambda)L] \quad (2)$$

The nonlinear transmissive intensity is

$$I_N(\lambda)=I(\lambda)exp[-\alpha_0(\lambda)L-\beta(\lambda)I(\lambda)L] \quad (3)$$

where L is the sample thickness, C is the ratio of the nonlinear excitation intensity to the linear excitation intensity.

From Eqs. (2) and (3), the ratio of the nonlinear transmissivity $T_N(\lambda)$ to the linear transmissivity $T_L(\lambda)$ can be written as

$$\eta(\lambda)=\frac{T_N(\lambda)}{T_L(\lambda)}=\frac{I_N(\lambda)}{CI_L(\lambda)}=exp[-\beta(\lambda)I(\lambda)L] \quad (4)$$

Therefore we have, for the nonlinear absorption coefficient,

$$\beta(\lambda)=-\frac{ln\eta(\lambda)}{I(\lambda)L} \quad (5)$$

For the intensity dependent third order nonlinear refractive process in the small signal regime, the refractive index can be written as

$$n(\lambda)=n_0(\lambda)+\triangle n(\lambda)=n_0(\lambda)+\gamma(\lambda)I(\lambda) \quad (6)$$

where $n_0(\lambda)$ is the linear refractive index and $\gamma(\lambda)$ is the nonlinear refractive index.

The change in the refractive index, $\triangle n(\lambda)$, can be obtained from the change in the absorption coefficient, $\triangle\alpha(\lambda)$, by the Kramers-Kronig transformation

$$\triangle n(\lambda)=\frac{1}{2\pi^2}P\int_0^\infty \frac{\lambda^2\triangle\alpha(\lambda')}{\lambda^2-\lambda'^2}d\lambda' \quad (7)$$

where P stands for the principal value of the integral.

Substituting Eqs. (1) and (6) into Eq. (7) and noting Eq. (5), we obtain for the nonlinear refractive index that

$$\gamma(\lambda)=-\frac{1}{2\pi^2 I(\lambda)L}P\int_0^\infty \frac{\lambda^2 ln\eta(\lambda')}{\lambda^2-\lambda'^2}d\lambda' \quad (8)$$

By Eq. (8) one can obtain the nonlinear refractive index from the linear and nonlinear transmission spectra and the laser intensity distribution on the frequency. Even though this is an indirect method of measuring the nonlinear refractive index, it has the advantage of providing correct values of $\gamma(\lambda)$ using a simple experimental setup. Especially, this method can give the nonlinear refractive index in a large wavelength range.

3. Experimental Results and Discussion

We measured the nonlinear refractive index for ZnSe-ZnS MQWs with the transmission spectrum method described above. The experimental setup is shown in Figure 1. The light source was from a tunable dye laser using coumarin 440 pumped by the nitrogen laser with a pulse duration of 10 ns and a repetition rate of 10 Hz. The calibrated neutral filters was used to limit the power of the beam passing through the sample. In measuring the linear transmission spectrum we used a very low-intensity excitation. However, in measuring nonlinear transmission a strong excitation was used. The intensity of excitation was varied by changing the neutral filters. In order to obtain nonlinear refractive index and nonlinear absorption coefficient, a nonlinear transmission spectrum must be measured in the small signal regime, otherwise, the two parameters obtained is not correct. The sample studied here was a ZnSe-ZnS MQWs with a total thickness of 1 μm grown by metal-organic chemical vapor deposition (MOCVD) on a transparent CaF_2 substrate and consisting of 100 periods.

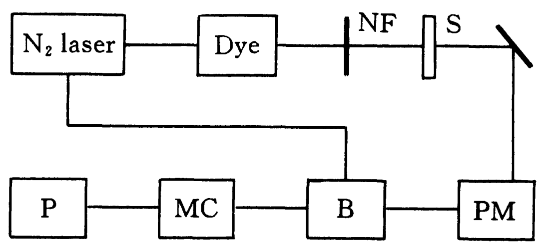

Fig. 1 Experimental setup for measuring the optical nonlinearities. Dye-tunable dye laser, NF-neutral filtters, S-sample, PM-photomutiplier, B-boxcar 4400, MC-microcomputer, P-printer.

Fig. 2 shows the transmission intensities of ZnSe-ZnS/CaF_2 MQWs as a function of wavelength for various incident intensities at 77 K. The curve labeled 1 is the linear transmission spectrum under a very low-power density excitation, the two observed absorption peaks around 441.3 nm and 435.5 nm correspond to the n=1 heavy-hole and n=1 light-hole excitons.[10] The compressive uniaxial component of the lattice mismatch strain in the ZnSe layers results in a splitting of the valence band degeneracy, shifting the heavy hole towards the conduction band with respect to the light hole. In the nonlinear transmission spectrum, curve labeled 2 in figure 2, both the exciton peaks are bleached and blue shifted slightly. When stronger excitation was used, the two exciton peaks gradually disappeared as the excitation intensity increased. The results indicate that the dominant nonlinear absorption saturation mechanism may be due to the phase

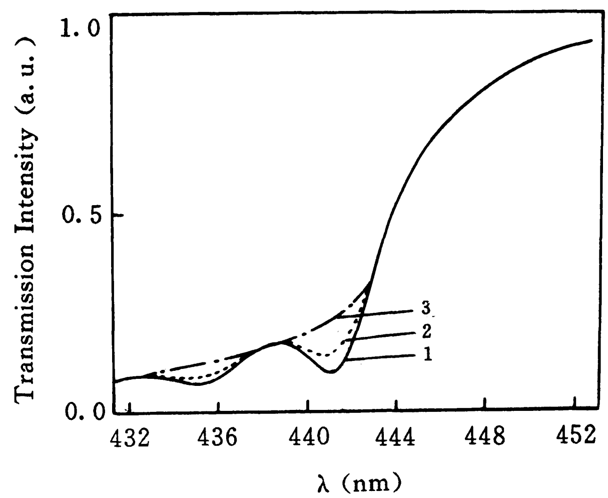

Fig. 2 Transmission spectra of ZnSe-ZnS/CaF_2 MQWs for various incident intensities at 77 K. (1)Corresponding to very low excitation intensity, (2), (3) corresponding to 6.0 and 17 kw/cm² excitation intentisies at λ=441.3 nm, respectively.

space filling of excitonic state.[11]

Using Eqs. (5) and (8), we calculated the nonlinear absorption coefficient and nonlinear refractive index from the transmission data. The results are

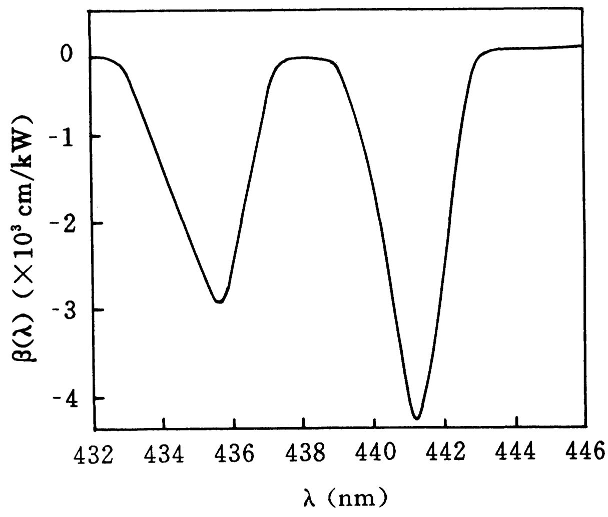

Fig. 3 Nonlinear absorption coefficient as a function of wavelength for ZnSe-ZnS/CaF_2 MQWs at 77 K.

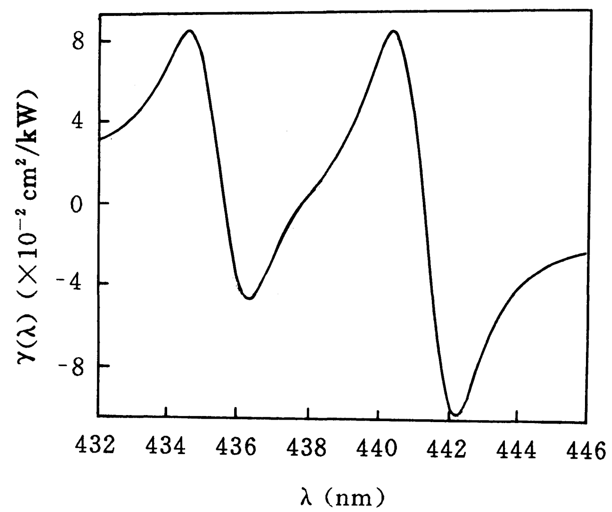

Fig. 4 Nonlinear refractive index as a function of wavelength for ZnSe-ZnS/CaF_2 MQWs at 77 K.

shown in Fig. 3 and Fig. 4. We see that the nonlinear absorption exhibited two peaks about λ=435. 5 nm and λ=441. 3 nm. The nonlinear refraction exhibited four peaks about λ=434. 6 nm, λ=436. 4 nm, λ=440. 4 nm and λ=442. 2 nm, where the first two peaks originate from λ=435. 5 nm nonlinear absorption peak, and the latest two peaks originate from λ=441. 3 nm nonlinear absorption speak. Namely, there are two nonlinear refraction peaks with contrary sign on two sides of each nonlinear absorption peak. A similar result has been obtained by peyghambarian.[6)]

4. Conclusion

We have derived the relation between the nonlinear refractive index and the ratio of nonlinear transmission to linear transmission, and shown how the magnitude and sign of the nonlinear absorption coefficient and nonlinear refractive index are determined from the transmission spectra data. Our method is simple and sensitive. It is expected that this method be a valuable tool in searching for highly nonlinear materials. We have applied this technique to study optical nonlinearities for ZnSe-ZnS/CaF_2 MQWs, and obtain the nonlinear refractive index as a function of wavelength.

Acknowledgements

This work is supported by the "863" High Technology Research Program in China, The National Natural Science Foundation of China, Laboratory of Excited State Process, Changchun Institute of Physics, Chinese Academy of Sciences, and the National Fundamental and Applied Research Projects of China.

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