Dispersion of nonlinear optical susceptibility of GaAs/Al_xGa_{1-x}As multiple quantum wells in the exciton region

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The dispersion of $\chi^{(2)}(2\omega)$ by the refelction of incident light from GaAs/Al_xGa_{1-x}As multiple quantum wells at room temperature with wavelength between 8000 and 9000 Å has been measured. The nonlinear susceptibility $|\chi_{14}^{(2)}(2\omega)|$ shows two pronounced peaks at 8436 and 8490 Å. It is suggested that one peak corresponds to the light-hole-exciton absorption and the other to the heavy-hole-exciton absorption.

I. INTRODUCTION

Dispersion measurements of the nonlinear optical susceptibilities for second-harmonic generation (SHG) in semiconductors have always been an interesting subject in nonlinear optics. There already exist several reports on the dispersion of nonlinear susceptibilities of semiconductors.[1–3] In that literature, authors found pronounced resonances in $\chi^{(2)}(2\omega)$ due to exciton absorption. Bethune, Schmidt, and Shen[2] showed that the nonlinear susceptibility $|\chi^{(2)}(2\omega)|$ has resonant structure when either ω or 2ω (or both) approaches the resonant peaks in the linear dielectric constant. Thus, in a given frequency range, the spectrum of $|\chi^{(2)}(2\omega)|$ should reveal more structure than the linear optical spectrum.

In the last decade, nonlinear properties of multiple quantum wells (MQW's) and superlattices (SL's) have attracted much attention. Many papers[4–7] have reported calculations and observations of strong third-order nonlinearities in GaAs/Al_xGa_{1-x}As MQW and SL structures arising from their large room-temperature excitonic absorptions. Among the phenomena related to excitonic behavior in GaAs/Al_x Ga_{1-x}As MQW structures such as exciton line saturation, optical bistability, the quantum confined Stark effect (QCSE), practical QCSE devices, so-called self-electro-optic-effect devices, and so on, have been demonstrated. In this paper we attempt to understand if the pronounced resonant dispersion due to the large room-temperature excitonic absorptions can be observed in the second-order nonlinear response of GaAs/Al_xGa_{1-x}As MQW structures. It is well known that the second-order optical nonlinearity is large only in noncentrosymmetric systems. Because of the symmetry of the system, quantum-well-type structures possess an inversion symmetry leading to the absence of second-order effects. Also, there is the fact that atoms at the surface of a dense medium are never at a position of inversion symmetry.[8] Therefore we consider here a reflected second-harmonic generation (SHG) from the epitaxy surface layers of GaAs/Al_xGa_{1-x}As MQW structure to study the $\chi_{14}^{(2)}(2\omega)$ dispersion behavior in which we are interested.

II. EXPERIMENTAL DESIGN

The room-temperature excitonic peaks (the lowest heavy-hole and light-hole excitons) of GaAs/Al_xGa_{1-x}As MQW structure, in principle, are in the wavelength range of 8000–9000 Å. Within this range, both the fundamental and harmonic waves are absorbed by the MQW samples. The method of reflected harmonic generation is therefore the best means of measuring the nonlinear susceptibility. In this case, the SHG in GaAs/Al_xGa_{1-x}As MQW structure originates from a volume restricted by the absorption depth of the second-harmonic light which is on the order of $\frac{1}{4}\lambda(2\omega)$.

In the following, the sample preparation, experimental setup, and SHG measuring procedure are reported.

A. Experimental arrangement

We used a pulsed tunable dye laser (with H_2 Raman cell) pumped by a Q-switched yttrium aluminum garnet (YAG) laser as the source of the fundamental beam in our experiments. The first-order Stokes output of this laser system was well polarized, 10 nsec long, with a peak power of 10–100 kW, having a linewidth of about 0.3 Å, a beam diameter of about 2 mm, and a tuning range from 8100 to 8800 Å. The overall experimental arrangement is shown in Fig. 1. An aperture was used to separate the first-order Stokes laser beam from others. After passing through an infrared filter (transparent from 0.725 to 1.095 μm) and a beam splitter, the laser beam was separated into two parts. One beam was directed through a Z-cut, 1.5-mm-thick quartz platelet, which generated the second harmonics used for normalization against possible laser fluctuations. The other beam was used for second-harmonics reflection measurements from the crystal samples. A 100-mm-focal-length lens was used to focus the laser beams on the sample and on the quartz platelet. Both the second-harmonic beams from the quartz and the sample were filtered, respectively, by a 5-cm-long cell of saturated $CuSO_4$ solution and an interference filter peaked at the harmonic wavelength (4000–4400 Å) to ensure that only the second-harmonic

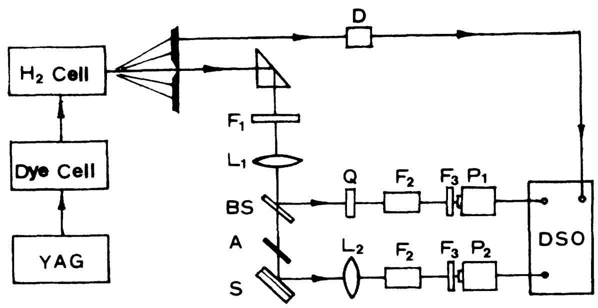

FIG. 1. Experimental setup: BS beam splitter; L_1, L_2, lenses; F_1, infrared filter (0.725–1.09 μm); F_2, $CuSO_4$, filter (5-cm cell); F_3, interference filter (4000–4400 Å); A, neutral attenuator; S, crystal sample; Q, normalization quartz; P_1, P_2, photomultipliers; D, diode; DSO, dual storage oscilloscope.

signals could be detected by the photomultipliers. The two output pulses from the photomultipliers were simultaneously recorded using a dual storage oscilloscope. Note that the intensity of the fundamental laser beam has to be controlled carefully to ensure that no surface damage is caused which may induce error signals coming from the plasma produced. Each measured datum was obtained by averaging the results of 20 laser shots.

B. Sample orientation and SHG

In our experiment the samples were grown using a computer-controlled molecular-beam-epitaxy system. The MQW structure consisted of ten periods, each containing 100-Å wells of GaAs and 100-Å barriers of $Al_{0.3}Ga_{0.7}As$ grown on a semi-insulating (001) GaAs substrate. Because the widths of the wells in our sample are on the order of a few tens of lattice constants, we can retain the $\bar{4}3m$ symmetry in our MQW sample orientation analysis.[9] The expression for $\chi_2(2\omega)$ for a crystal with $\bar{4}3m$ symmetry is[10]

$$\chi_{14}^{(2)}(2\omega)=A\int_{\mathrm{BZ}}d^3K\sum_{l',l}\left[\frac{Q_{l'l}^{(1)}(\mathbf{k})}{\omega^2-\omega_{l'l}^2}+\frac{Q_{l'l}^{(2)}(\mathbf{k})}{4\omega^2-\omega_{l'l}^2}\right], \quad (1)$$

where A is a constant and $\omega_{l'l}=(1/\hbar)(E_{l'}-E_l)$. The summation is taken over the band indices, and the integration over the Brillouin zone. The Q's are real and independent of frequency. Following Bloembergen and Pershan's analysis,[11] when the laser polarization is parallel to the $[1\bar{1}0]$ direction in a (001) face the second-harmonic polarization has the nonzero component $P_z^{\mathrm{NLS}}(2\omega)=\chi_{14}^{\mathrm{NL}}E_x(\omega)E_y(\omega)$. The nonlinear polarization in this case is in the plane of reflection. When the incident angle $\theta=45°$, and the rotation angle ϕ about the Z axis of the crystal is equal to zero (as in our case), we deduce the second-harmonic reflected electric field polarized in the plane of reflection to be

$$E_{\parallel}^{R}(2\omega)=\frac{8^{1/2}\pi\chi_{14}^{(2)}(2\omega)}{[\alpha(\omega)+\alpha(2\omega)][\alpha(2\omega)+\epsilon(2\omega)]}E_i^2(\omega)\ , \quad (2)$$

where $\alpha(\omega)=[2\epsilon(\omega)-1]^{1/2}$, $\alpha(2\omega)=[2\epsilon(2\omega)-1]^{1/2}$, and $\epsilon(\omega)$ and $\epsilon(2\omega)$ are the linear dielectric constants of the $GaAs/Al_xGa_{1-x}As$ MQW samples at the fundamental and double frequencies, respectively.

We rewrite Eq. (2) as

$$|E_{\parallel}^{R}(2\omega)|^2=A(\omega)|\chi_{14}^{(2)}(2\omega)|^2E_i^4(\omega)\ , \quad (3)$$

where

$$A(\omega)=\frac{8\pi^2}{[\alpha(\omega)+\alpha(2\omega)]^2[\alpha(2\omega)+\epsilon(2\omega)]^2}\ .$$

The Z cut was chosen for the monitoring quartz crystal so that one does not have to worry about the orientation of the laser polarization with respect to the crystalline axis. Moreover, a Z cut quartz is far from the phase-matching condition, thus accidental misalignment of the monitoring system would not affect the second-harmonic conversion. In our experimental wavelength range which is far from any absorption in quartz it is reasonable to assume that the dispersion of the nonlinear susceptibility of the quartz can be ignored. Therefore the second-harmonic output from the quartz is proportional to

$$|E_Q^T|^2=B\chi_{11}^{(2)}E_j^4(\omega)\ , \quad (4)$$

where B is a constant corresponding to the linear optical constants of the quartz crystal.

C. Experimental procedure

We measured the second-harmonic output $P_s(2\omega)$ generated from the $GaAs/Al_{0.3}Ga_{0.7}As$ MQW sample mentioned above by reflection and the second-harmonic output $P_Q(2\omega)$ from the quartz platelet by transmission. At each laser frequency the results were averaged over 20 laser pulses. According to Eqs. (3) and (4) and comparing the ratio of the two second-harmonic intensities we have

$$|\chi_{14}^{(2)}(2\omega)|/|\chi_{11}^{(2)}|\propto C(\omega)[P_S(2\omega)/P_Q(2\omega)]^{1/2}\ , \quad (5)$$

where $C(\omega)=[A(\omega)]^{-1/2}$, corresponding to the linear dielectric constants of the MQW sample. Using the theoretical fitting to the curve of experimental reflectivity measured in the range of ω and 2ω, we calculated $C(\omega)$, which on substitution into Eq. (5) gave the dispersion of $\chi_{14}^{(2)}(2\omega)$ of the sample.

III. RESULTS AND DISCUSSION

Curve (a) of Fig. 2 shows the experimental results of $|\chi_{14}^{(2)}(2\omega)|/|\chi_{11}^{(2)}|$ in the wavelength range of 8100 to 8900 Å for the $GaAs/Al_{0.3}Ga_{0.7}As$ MQW sample. It can be seen from curve (a) of Fig. 2 that the measured nonlinear susceptibility $|\chi_{14}^{(2)}(2\omega)|$ shows two pronounced peaks at 8436 and 8490 Å, respectively. Comparing with the fluorescence spectrum of the sample shown in Fig. 3, the positions and the interval between the two structures in the dispersion of $|\chi_{14}^{(2)}(2\omega)|$ suggest that one peak (8436 Å) corresponds to the light-hole-exciton absorption and the other one (8490 Å) arises from the heavy-hole-exciton absorption. Under these circumstances the first term inside the large parentheses of Eq. (1) ought to predominate entirely, that is, one-photon single resonances govern the behavior of $|\chi_{14}^{(2)}(2\omega)|$ in the neighborhoods of the light- and the heavy-hole-exciton energy. As a comparison, we also measured the dispersion of $|\chi_{14}^{(2)}(2\omega)|$ of pure GaAs

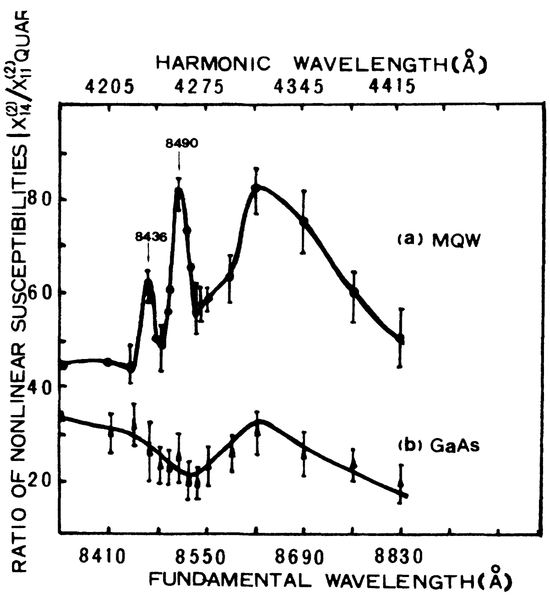

FIG. 2. Experimental ratios of nonlinear susceptibilities $|\chi_{14}^{(2)}(\text{sample})|/|\chi_{11}^{(2)}(\text{quartz})|$ in the wavelength range of 8100–8900 Å: curve (a), for the GaAs/$Al_{0.3}Ga_{0.7}As$ MQW

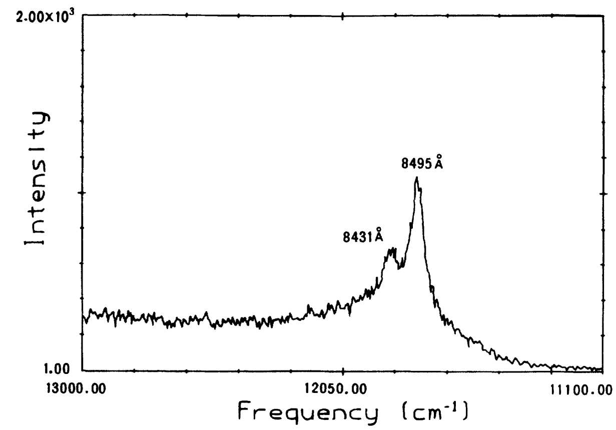

FIG. 3. Fluorescence spectrum of the GaAs/$Al_{0.3}Ga_{0.7}As$ MQW sample at room temperature: the heavy-hole-exciton peak is clearly evident (at 8495 Å), but the light-hole-exciton peak (8431 Å) can hardly be distinguished from the background.

which has the same orientation and the same polished surface as a GaAs substrate of the MQW sample under the same experimental conditions. As shown in curve (b) of Fig. 2, no resonant structure in $|\chi_{14}^{(2)}(2\omega)|$ was found in this pure GaAs sample in the range of the two excitonic absorption peaks mentioned above.

It can be seen clearly that in both curves (a) and (b) of Fig. 2 there is an obviously broader peak at about 8671 Å. The energy of this peak is equal to one-half of the E_1 gap (2.89 eV) of GaAs crystal.[12] The transition associated with the E_1 energy corresponds to the extended state resonance which is not confined to the GaAs quantum wells. Thus this broader peak structure of $|\chi_{14}^{(2)}(2\omega)|$ in both the GaAs/$Al_{0.3}Ga_{0.7}As$ MQW and the pure GaAs samples is probably due to two-photon single resonance which is given by the second term in Eq. (1).[13]

It is worthy to note that in the fluorescence spectrum of the MQW sample (see Fig. 3) we can clearly observe the heavy-hole-exciton absorption peak (8495 Å). However, we can barely distinguish the light-hole-exciton peak (8431 Å), which has only a very weak structure, from the background. In the dispersion spectrum of the MQW sample [curve (a) of Fig. 2], two resonant structures which arise from the heavy hole and the light hole excitons are clearly displayed. This means that the spectrum of $|\chi_{14}^{(2)}(2\omega)|$ can, in principle, yield more detailed information about the band structure of material.

In conclusion, we have measured for the first time the dispersion of $|\chi_{14}^{(2)}(2\omega)|$ by reflection from a GaAs/$Al_{0.3}Ga_{0.7}As$ MQW structure at room temperature in the wavelength range of 8100–8900 Å. The nonlinear susceptibility $|\chi_{14}^{(2)}(2\omega)|$ shows two pronounced resonant peaks at 8436 and 8490 Å. In comparison with the $|\chi_{14}^{(2)}(2\omega)|$ spectrum of pure GaAs and with the fluorescence spectrum of the MQW sample, the results can clearly be interpreted in terms of the large heavy hole (8495 Å) and light hole (8431 Å) room-temperature excitonic absorptions, respectively.

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