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Optical second-order susceptibility of GaAs/$Al_xGa_{1-x}As$ asymmetric coupled-quantum-well structures in the exciton region

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We have measured the dispersion of $\chi^{(2)}$ of GaAs/$Al_xGa_{1-x}As$ asymmetric coupled-quantum-well (ACQW) structures in the region of room-temperature excitonic resonances. The spectrum of $\chi^{(2)}$ has sharp and strong peaks including the transitions which are allowed by selection rule in a symmetric quantum well (SQW) as well as the transition which is forbidden in a SQW. The peak value of $\chi^{(2)}_{14}(2\omega)$ of the ACQW which we measured is ten times larger than that of bulk GaAs.

I. INTRODUCTION

The second-order nonlinearities of quantum wells (QW) and superlattices (SL), as well as the third-order ones, are of great practical significance in the fields of optical modulation, optical communication, etc. As the existence of excitations strongly enhances the third-order susceptibility $\chi^{(3)}$ of SL's and multiple QW's (MQW's), we expect it also to affect $\chi^{(2)}$, the second-order susceptibility of these materials. We have observed the dispersion of $\chi^{(2)}_{14}(2\omega)$ of the symmetric quantum well in the exciton region by reflected second-harmonic generation (SHG).[1] In this paper we present our latest results on the study of $\chi^{(2)}$ of a GaAs/$Al_xGa_{1-x}As$ asymmetric coupled-quantum-well (ACQW) structure.

In a symmetric QW, the structural contribution to SHG is entirely eliminated due to its centrosymmetrical structure, so the reflected second-order nonlinear response of the QW in Ref. 1 is due to the surface effect, in addition to the excitonic modulation at room temperature. The value of $\chi^{(2)}_{14}$ was observed to be enhanced several times due to this modulation in comparison with that in the bulk GaAs.

Khurgin[2] predicted theoretically that there should be a large second-order susceptibility in a system of GaAs/$Al_xGa_{1-x}As$ asymmetric coupled QW's. He calculated the $\chi^{(2)}_{15}$ (same as the $\chi^{(2)}_{14}$ in our case) in an ACQW structure where the SHG frequency 2ω is close to the band gap. The value of $\chi^{(2)}_{15}$ is less than the $\chi^{(2)}_{31}$ of $LiNbO_3$ which is about $\frac{1}{7}$ of bulk GaAs. We have relatively measured the second-order nonlinear susceptibility $\chi^{(2)}_{14}$ and the dispersion relation of an ACQW structure, in which the fundamental frequency ω is in the region of room-temperature excitonic resonance (i.e., ω is close to the band gap). We found that the peak value of $\chi^{(2)}_{14}$ for GaAs/$Al_xGa_{1-x}As$ ACQW is much larger than that of bulk GaAs in the same region.

II. EXPERIMENT

Due to the strong absorption of both harmonic and fundamental frequencies within the GaAs/$Al_xGa_{1-x}As$ QW near the exciton region, reflected detection was used in our experiment. The pump source was a pulsed tunable dye laser driven by a Q-switched yttrium aluminum garnet (YAG) laser. The tunable range was about 6500–7500 Å. The experimental arrangement is shown in Fig. 1. The laser beam was separated into two parts. One beam was directed through the quartz crystal Q_1 (Z cut, 1.5 mm thick), which generated the second harmonic used for normalization against possible laser fluctuations. The other beam was used to generate second harmonic either by reflection from the ACQW sample or by transmission through a similar quartz crystal Q_2 (dotted path in Fig. 1). The second harmonic signal from this quartz Q_2 was needed for calibration of the nonlinear susceptibility of the sample. Each measured datum was obtained by averaging the results of over 30 laser shots using a 20-8 instantaneous recorder-computer system.

The sample used has ten periods of ACQW, each with 14 Å of GaAs well, 14-Å $Al_{0.5}Ga_{0.5}As$ barrier, 17-Å GaAs well, and 35-Å $Al_{0.5}Ga_{0.5}As$ barrier, respectively. Thus in the same period, there is strong electron and hole tunneling between the two coupled QW's. This structure is obviously noncentrosymmetric and should possess structural $\chi^{(2)}$.

During the experiment, we simultaneously measured the SHG output $P_s(2\omega,p_s,A_s)$ of the ACQW sample by reflection and $P_{Q_1}(2\omega,p_{Q_1}A_{Q_1})$ of the Q_1 quartz platelet by transmission. The SHG output $P'_{Q_2}(2\omega,p'_{Q_2},A'_{Q_2})$ of the Q_2 quartz platlet and $P'_{Q_1}(2\omega,p'_{Q_1},A'_{Q_1})$ of the Q_1 were also measured both by transmission, simultaneously. The incident angle θ on the ACQW sample was 45°. In the above expressions p and p' are the laser powers, and A and A' the effective cross sections of the beam. For reflective SHG with $\theta=45°$, we have

$$P_s(2\omega,p_s,A_s)=\frac{64\pi^3|\chi^{(2)}_{14}(2\omega)|}{Cn_0[\alpha_s(\omega)+\alpha_s(2\omega)]^2[\alpha_s(2\omega)+\epsilon_s(2\omega)]^2}\frac{P_s^2(\omega)}{A_s}\ , \qquad (1)$$

where C is the velocity of light in air, $n_0=1$, $\alpha_s(2\omega)=[2\epsilon_s(2\omega)-1]^{1/2}$, $\alpha_s(\omega)=[2\epsilon_s(\omega)-1]^{1/2}$, and $\epsilon_s(\omega)$ is the dielec-

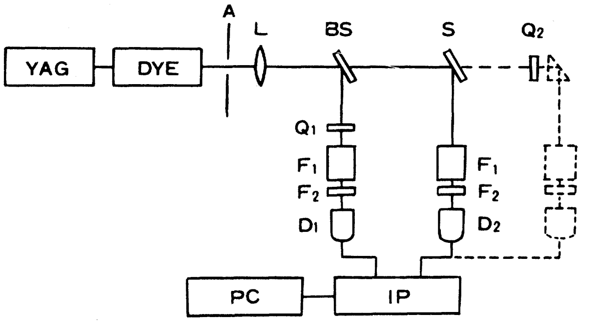

FIG. 1. Experimental setup. A, aperature; BS, beam splitter, with $R=35\%$ for an incident angle of 45°; L, lens; S, sample; Q_1, Q_2, Z-cut quartz platelets; F_1, 5-cm cell of saturated $CuSO_4$ solution; F_2, ultraviolet filter (3250–4000 Å); D_1, D_2, photomultipliers, IP, instantaneous processor, PC, computer.

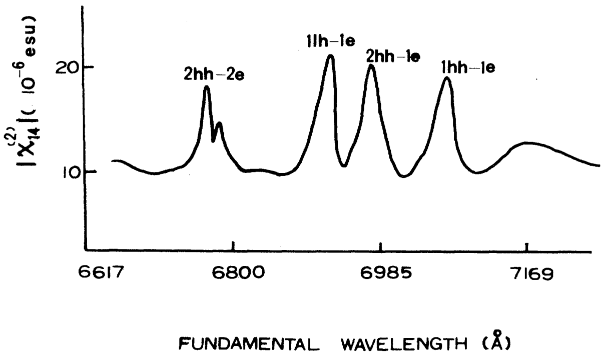

FIG. 2. Experimental values of nonlinear susceptibilities $|\chi_{14}^{(2)}|$ of the ACQW sample in the range 6600–7300 Å.

tric constant of the sample. For transmission, we have

$$P_Q(2\omega, p_Q, A_Q)=\frac{32\pi^3\omega^2}{c^3\epsilon_s(\omega)\sqrt{\epsilon_s(2\omega)}}|\chi_{11}^{(2)}(2\omega)|^2 Z^2\frac{\sin^2(\Delta kZ/2)}{(\Delta kZ/2)^2}\frac{p_Q^2(w)}{A_Q}\ , \tag{2}$$

where Z is the thickness of the platelet, and $\Delta k=[n_Q(2\omega)-n_Q(\omega)]\omega/c$. Rewriting Eqs. (1) and (2):

$$P_s(2\omega,p_s,A_s)=C_s(\omega)p_s^2(\omega)/A_s,\quad P_Q(2\omega,p_Q,A_Q)=C_Q(\omega)p_Q^2(\omega)/A_Q\ .$$

So

$$P_s/P_{Q_1}=(C_sA_{Q_1}/C_{Q_1}A_s)[(1-R)/R]^2,\quad P_{Q_2}/P_{Q_1}=(C_{Q_2}A_{Q_1}/C_{Q_1}A_{Q_2})[(1-R)/R]^2\ ,$$

where R is the reflectivity of the beam splitter. Setting $A_s=A_{Q_2}$, we obtain

$$\frac{P_s/P_{Q_1}}{P_{Q_2}/P_{Q_1}}=\frac{C_s(\omega)}{C_{Q_2}(\omega)}=\gamma\ . \tag{3}$$

Note that the ratio γ is independent of the light power and the cross section. In addition, it is reasonable to set $\sin^2(\Delta kZ/2)=\frac{1}{2}$, which indicates the averaging effect of the "Maker fringe" factor over the laser bandwidth and random jitter of the laser frequency.[3]

We then combine Eqs. (1) and (2) with (3),

$$|\chi_{14}^{(2)}(2\omega)|_s=\left[\frac{4\gamma}{n_Q(2\omega)}\right]^{1/2}\left|\frac{[\alpha_s(\omega)+\alpha_s(2\omega)][\alpha_s(\omega)+\alpha_s(2\omega)]}{n_Q(\omega)[n_Q(2\omega)-n_Q(\omega)]}\right||\chi_{11}^{(2)}(2\omega)|\ , \tag{4}$$

where γ can be obtained easily from the experimental data, and the linear optical constants of the ACQW sample are calculated by fitting the experimental reflected spectrum of the sample using an appropriate method, e.g., Ref. 4. Equation (4) then gives the dispersion of $|\chi_{14}^{(2)}(2\omega)|$ of the ACQW sample.

III. RESULTS AND DISCUSSION

The incident wavelength range was 6600–7300 Å, which covered the room-temperature excitonic resonances of the sample. Figure 2 shows the experimental results of the $|\chi_{14}^{(2)}(2\omega)|$ dispersion spectrum of the $GaAs/Al_xGa_{1-x}As$ ACQW.

There are very rich structures appearing in the dispersion curve. Comparing with the case of symmetric QW's, here we observe not only the resonant dispersion peaks arising from allowing interband transitions of the heavy holes and the light holes (1hh-1e; 2hh-2e; 1lh-1e; 2lh-2e), but also the forbidden interband transition (2hh-1e) which is broken by the asymmetry structure of the sample. In addition, we have relatively measured the value of $\chi_{14}^{(2)}$ of our ACQW's by means of quartz calibration. Taking $\chi_{11}^{(2)}=0.82\times1.50\times10^{-9}$ esu, we obtain the peak value of $\chi_{14}^{(2)}$ to be 2×10^{-5} esu, which is ten times that of bulk GaAs. This experimental result is much larger than Khurgin's theoretical value in which 2ω is in the band-gap range.[2]

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