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SECOND-HARMONIC GENERATION IN ZnCdSe/ZnSe ASYMMETRIC DOUBLE QUANTUM WELLS

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Second-harmonic generation in ZnCdSe/ZnSe asymmetric double quantum wells (ADQW) was observed using the Maker fringe technique. It is due to the noncentrosymmetric characteristics of ADQW, which make $X^{(2)}$ not equal to zero. The birefringence in ADQW was also observed by finding the point of phase match between fundamental wave and second harmonic.

Second-harmonic generation (SHG) is one of the important second order nonlinear optical effect, which has been studied widely. As we know, in centrosymmetric materials, second-order susceptibility ($X^{(2)}$) is zero, which induce that the SHG can not be observed, so in the past, studies were concentrated on noncentrosymmetric materials, of which $X^{(2)}$ is not zero. However now, the developed methods of epitaxy such as MBE and MOCVD make it possible to prepare super thin film in a few lattice constants range, which allows us growing alternating asymmetric thin layers of semiconductors with different material in order to constitute a kind of noncentrosymmetric material. By this method, many kinds of noncentrosymmetric materials can be manufactured using centrosymmetric materials. In this paper, we report the observation of SHG in ZnCdSe/ZnSe asymmetric double quantum wells (ADQW) by Maker fringe technique. Although SHG has been studied in asymmetric quantum wells [1, 2], all of them were studied by intersubband resonant transitions and the wavelength of SHG was in infrared range. Here we studied the SHG in green range without intersubband transitions.

The ZnCdSe/ZnSe ADQW structure used in the experiment was grown by low-pressure MOCVD on GaAs substrate at 350°C. The ADQW structure constitutes a 1 μm ZnSe buffer layer on substrate, a five-period 3 nm $Zn_{0.72}Cd_{0.28}Se$ layer/2.5 nm ZnSe layer/7 nm $Zn_{0.72}Cd_{0.28}Se$ layer ADQW structure separated by a 30 nm ZnSe layer and a 60 nm ZnSe cap layer. The sample was excited by a 1064 nm line of a Nd : YAG laser working at 1 Hz at room temperature and SHG signal passed through the 532 nm filter and then was accepted by a photometer connected to a computer. The reflected scheme was used in the measurement and the incident light was focused on 1 mm^2.

Figure 1 shows the dependence of the SHG signal intensity on the incident angle. From Fig. 1, we find that the SHG signal can be resolved into two parts, one is the part like sinusoid on the whole range; the other one is that at $\theta = 20$ degrees there is a sharp increasing. In $Zn_{0.72}Cd_{0.28}Se/ZnSe$ ADQW structure, the absorption edge of material is larger than the energy of 532 nm light but the difference between intersubbands of the ADQW is surely smaller than the energy of 532 nm light and 1064 nm light. Therefore we believe that the SHG is not caused by intersubband resonant transitions and also can assume that the incident light is not lost in ADQW structure. According to nonlinear optical theory, under the condition of dephasing, the amplitude intensity of the second harmonic can be expressed as [3]:

$$\epsilon(2\omega, z) = \epsilon(\omega, 0)\frac{\sin\left(\frac{\Delta k}{2}z\right)}{\left(\frac{\Delta k}{2}\right)L_{sh}} \tag{1}$$

where z is the effective thickness of material, $\epsilon(\omega, 0)$ is the amplitude intensity of fundamental waves, Δk is the dephasing between fundamental waves and second harmonic waves. $L_{sh} = [\omega^2(k_{2\omega}c^2)/|\chi_{2\omega}|\epsilon(\omega, 0)]^{-1}$ is the characteristic length, ω is fundamental wave frequency $\chi_{2\omega}$ is second-harmonic susceptibility, c is velocity of light in free space. In our discussion, we neglect the second harmonic generated from reflection of the

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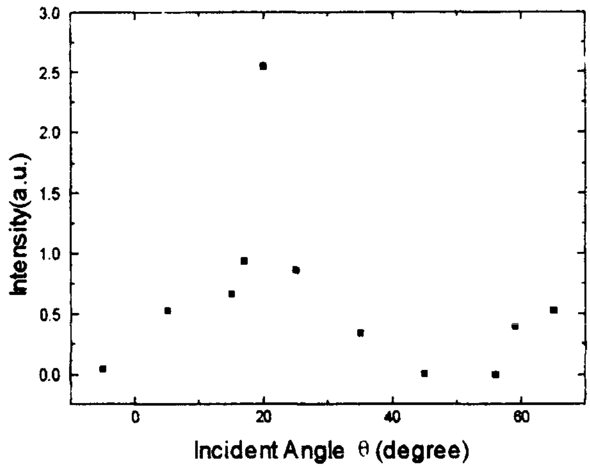

Fig. 1. The dependence of SHG intensity on incident angle.

incident light on the interface between the ZnSe buffer and GaAs layer, so in equation (1), $z = d/\cos\theta$, where d is the thickness of the material. From equation (1), we find that two parameters are varied with θ, one is Δk, the other is z and the total effect is the amplitude intensity of second harmonic $\epsilon(2\omega, z)$ shows sine vibration with θ. For the reason that the ADQW structure is very thin (total thickness is less than 100 nm), the varying range of z is narrow which makes the period of the amplitude large.

As $\theta = 20°$, the sharp increase of $\epsilon(2\omega, z)$ is attributed to phase match between the incident light and SHG [4]. Under the condition of phase match $\Delta k = 0$, the amplitude of SHG can be expressed as

$$\epsilon(2\omega, z) = \epsilon(\omega, 0)\sec h\left(\frac{z}{L_{sh}}\right). \qquad (2)$$

Equation (2) shows that $\epsilon(2\omega, z)$ would be dominated by z, which means that the thicker the ADQW structure is, the stronger the SHG intensity will be. As discussed above, the ADQW structure studied is very thin, which limits the intensity of SHG. Therefore if increasing the thickness of the ADQW structure, the intensity of SHG will become stronger. From the phase match condition $\Delta k = 0$, we deduce that $2k_1 = k_2$ and then $n_1 = n_2$, where k_1, n_1, k_2, n_2 are the constants of spread and index of refraction of incident light and SHG respectively. On the condition of normal chromatic dispersion, n changes with ω, which mean it is impossible to meet condition $n_1 = n_2$, so there must exist birefringence characteristic in ADQW structure which can compensate the normal chromatic dispersion effect. Although we don't know much about the refraction of the ADQW structure, we can confirm that there must be the point ($\theta = 20$ degree), which make $n_{1o(e)} = n_{2e(o)}$, where n_o, n_e are refractive index of ordinary and extraordinary ray respectively.

From the changing of the amplitude intensity of SHG $\epsilon(2\omega, z)$, we also can draw the conclusion that there exists the birefringence characteristic. From the experiment result, we obtain that the two maximums of amplitude intensity of the SHG are not equal and the second one (at about $\theta = 65$ degree) is about half of the first one (at about $\theta = 20$ degree). If there is no birefringence in ADQW structure, the Δk will not change with the incident angle θ and then from equation (1), we know the maximum of $\epsilon(2\omega, z)$ is a constant equaled to $[2\epsilon(\omega, 0)]/(\Delta k L_{sh})$. Obviously, this does not tally with the experimental result. But if there is birefringence, Δk will be changed by changing the incident angle θ, so the maximum of $\epsilon(2\omega, z)$ is not a constant, but changed with Δk and if far away from the point that phase match, the Δk will increase and the maximum of $\epsilon(2\omega, z)$ will decrease, which is what we see from Fig. 1.

According to equation (1), under the condition of dephase, the period of vibration of the $\epsilon(2\omega, z)$ is given by $T = (4\pi)/\Delta k$. Due to the birefringence in ADQW, k changes with θ and then T will be decreased on increasing Δk. From experimental results we really find that the half width of the second maximum is smaller than that of the first maximum.

In ZnSe epitaxy layer and normal ZnCdSe/ZnSe multiple quantum well structure, we did not find any SHG, therefore we contribute the SHG observed here to the noncentrosymmetric characteristic of the ADQW structure. As we know, ZnSe is zincblende structure and normal ZnCdSe/ZnSe multiple quantum well structure grown on a thick ZnSe buffer is also zincblende structure and they all belong to T_d group and have the centrosymmetric characteristic which induces that $X^{(2)}$ is zero. However, for ZnCdSe/ZnSe ADQW structure which still belong to the zincblende structure, due to the different kinds of materials and the strain between the well layer and barrier layer, the centrosymmetric is changed and there is no centrosymmetric characteristic in the range of a few lattice constants. Although we didn't know how it would effect the index of refraction, we can be sure that $X^{(2)}$ is not zero.

In conclusion, with the developed epitaxy method, the ZnCdSe/ZnSe ADQW structure was prepared, which showed a noncentrosymmetric characteristic and the second-harmonic generation was observed using the Maker fringe technique because $X^{(2)}$ is not zero. That means the noncentrosymmetric material could be manufactured using centrosymmetric material by controlling the preparation conditions properly. We hope these results will be helpful on the material preparation and device design.

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