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# Second-harmonic generation from a GaAs/$Al_{1-x}Ga_x$As asymmetric quantum-well structure

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We report the observation of second-harmonic generation (SHG) in reflection from a GaAs/$Al_xGa_{1-x}$As asymmetric quantum-well (AQW) structure and a GaAs(001) face. SHG intensities are measured as a function of sample azimuthal angle for several polarization configurations using a pulsed 775-nm fundamental beam. Isotropic contributions to the *p*-polarized SHG intensity are found for both samples, which provides clear evidence of SHG arising from both the AQW structure and the GaAs(001)/oxide buried interface. The experimental results are consistent with a strong SHG contribution from the AQW structure with magnitude close to that of the bulk contribution; this interpretation is supported by previous theoretical predictions. Given the magnitude of the second-harmonic susceptibility for an AQW, strong SHG enhancement for visible and/or near-infrared light by full implementation of quasi-phase-matching is feasible.

The large optical nonlinearities arising from semiconductor heterojunctions and quantum wells (QW's), together with their possible related device applications, have attracted a great deal of interest in recent years. Among these are a variety of interesting and potentially useful second-order nonlinear effects, such as second-harmonic generation (SHG), difference-frequency generation (DFG), and parametric oscillation. In the electric dipole approximation, the existence of these second-order effects requires a noncentrosymmetric structure. It has been proposed that the second-harmonic (SH) susceptibility $\chi^{(2)}$ can be enhanced by two orders of magnitude over that arising from bulk materials using either intraconduction[1,2] or intravalence[3,4] subband transitions in asymmetric quantum wells (AQW's). The prediction involving intraconduction subband transitions was subsequently verified experimentally by several groups.[2,5] In these schemes, the fundamental wavelength required to achieve the double-resonance condition falls in the far-infrared region ($\gtrsim 5\ \mu$m). Recently, the possibility of an enhancement in the DFG susceptibility by $10-10^4$ times over the bulk value was proposed using similar structures.[6,7] On the other hand, interband transitions have to be utilized for SHG in the near-infrared and visible wavelength region. The SH susceptibility of AQW's in this wavelength range is generally predicted to be comparable to that of bulk media[8–10] due to the absence of a double-resonance enhancement. This prediction, however, if verified, permits the possibility of efficient SHG devices by the further employment of quasi-phase-matched (QPM) AQW domains.[4,10,11] Unfortunately, to the best of our knowledge there has been no report on visible and near-infrared SHG in AQW's. Only Lue, Lo, and Tzeng[12] have attempted to measure SHG from a symmetric QW and have confirmed the anticipated result that SHG from such an unbiased well is negligible compared to its bulk counterpart.

In this paper we report on SHG measured in reflection in air from a GaAs/$Al_xGa_{1-x}$As multiple AQW sample and from a GaAs(001) wafer. We used a 775-nm fundamental beam to suppress intentionally the bulk SH contribution by strong SH absorption. Our results provide strong evidence for the existence of a sizable SH contribution from the AQW structure and demonstrate the use of SHG in reflection as a sensitive probe of heterostructures of noncentrosymmetric semiconductors.[13]

We now consider SHG in reflection from noncentrosymmetric media in general. The SH field at the surface, $\mathbf{E}^{(2\omega)}$, excited by an incident electric field $\mathbf{E}^{(\omega)}$, can be written as[14]

$$\mathbf{E}^{(2\omega)} = \left[ \frac{i\omega\varepsilon_0}{c} \right] \mathbf{F}(2\omega)\cdot\{\chi_s^{(2)} + \chi_b^{(2)} L_b^{\text{eff}} + \chi_{\text{QW}}^{(2)} L_{\text{QW}}^{\text{eff}}\} :\{\mathbf{f}(\omega)\mathbf{f}(\omega)\mathbf{E}^{(\omega)}:\mathbf{E}^{(\omega)}\} \ . \quad (1)$$

Anticipating later results, magnetic dipole and electric quadrupole contributions are neglected in this equation. $\chi_s^{(2)}$ and $\chi_b^{(2)}$ are the surface and bulk SH susceptibility tensors; $\chi_{\text{QW}}^{(2)}$ describes the response of the AQW structure, and $\mathbf{f}(\omega)$ and $\mathbf{F}(2\omega)$ are the Fresnel factor matrices for the incident and SH fields, respectively. $L_b^{\text{eff}} = [\alpha_{2\omega}/2 + i\Delta k_z]^{-1}$ is the complex effective active length for bulk SHG, where $\alpha_{2\omega}$ and $\Delta k_z$ are the SH ab-

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sorption coefficient and phase mismatch (caused by dispersion), respectively.[14] In the same fashion, we also define $L_{\mathrm{QW}}^{\mathrm{eff}}$ as the effective active length for SHG from QW's, which depends on the active length of a single AQW, the SH absorption, and the phase difference of SH waves generated from different AQW's. It is clear that a relatively strong AQW contribution may be obtained by using, for instance, a short fundamental wavelength resulting in a large $\alpha_{2\omega}$ and therefore small $L_b^{\mathrm{eff}}$. Alternatively, $L_{\mathrm{QW}}^{\mathrm{eff}}$ can be increased by spacing opposite-sign AQW's, separated by the SH coherence length, $l_c(=\pi/\Delta k_z)$, such that a QPM scheme is realized.[4,10] Both these approaches are employed simultaneously in our experiment. One should bear in mind that strong SH absorption ensures that only shallow AQW's are being sampled. Therefore this method facilitates observations of SHG contributions from clean surfaces and shallow buried interfaces.[15]

For crystalline materials grown on GaAs(001) substrates, the dependence of the SH intensity $I_{g,h}^{(2\omega)}$ on sample azimuthal angle $\psi$ is given by[16,17]

$$I_{g,p}^{(2\omega)}(\psi) \propto |c_{g,p}^{(0)}+c_{g,p}^{(2)}\cos(2\psi)+c_{g,p}^{(4)}\cos(4\psi)|^2 \; , \tag{2a}$$

$$I_{p,s}^{(2\omega)}(\psi) \propto |c_{p,s}^{(2)}\sin(2\psi)+c_{p,s}^{(4)}\sin(4\psi)|^2 \; , \tag{2b}$$

where $g$ and $h$ are the fundamental and SH polarizations, respectively. It can be seen that the coefficients $c_{g,h}^{(m)}$ give rise to $m$-fold rotational symmetry in $E_{g,h}^{(2\omega)}(\psi)$, while $c_{g,p}^{(0)}$ represents an isotropic contribution. $c_{g,p}^{(0)}$ contains the contributions from both the interfacial dipole mechanism and the bulk quadrupole mechanism, while $c_{g,h}^{(4)}$ contains the contribution of the bulk quadrupole mechanism only. Both interfacial and bulk dipole mechanisms contribute to $c_{g,h}^{(2)}$. For GaAs and $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ crystals ($\bar{4}3m$ symmetry), where the only nonzero $\chi_b^{(2)}$ element is $\chi_{14}^{(2)}$ $(=\chi_{xyz}^{(2)})$, the bulk contribution to the $c_{g,h}^{(2)}$ coefficient, $c_{g,h}^{(2),\mathrm{bulk}}$, is given by[17]

$$c_{p,s}^{(2),\mathrm{bulk}} = -2f_c f_s \chi_{14}^{(2)} L_b^{\mathrm{eff}} \; , \tag{3a}$$

$$c_{s,p}^{(2),\mathrm{bulk}} = F_s \chi_{14}^{(2)} L_b^{\mathrm{eff}} \; , \tag{3b}$$

$$c_{p,p}^{(2),\mathrm{bulk}} = (2F_c f_c f_s - F_s f_c^2)\chi_{14}^{(2)} L_b^{\mathrm{eff}} \; , \tag{3c}$$

where $f_s, f_c$ and $F_s, F_c$ are the Fresnel factors defined in Refs. 16 and 17. The AQW symmetry class is tetragonal. Because the $z$ direction is different from $x$ and $y$ in the AQW, one has the following relation for the $\chi_{\mathrm{QW}}^{(2)}$ elements: $\chi_{14,\mathrm{QW}}^{(2)}=\chi_{25,\mathrm{QW}}^{(2)}\neq\chi_{36,\mathrm{QW}}^{(2)}$. This, however, does not alter the phenomenology of the SH sources described in Eq. (2). We also note that, in general, the $c_{g,h}^{(m)}$ are complex numbers, reflecting the complex nature of $\chi^{(2)}$-tensor elements, the Fresnel factors, and the phase differences between the different SHG sources.

The AQW sample used in our experiment is composed of two-step compositional AQW's. AQW's of this kind have been discussed thoroughly elsewhere.[4,10] It has been shown that the double-resonance enhancement condition (for SHG) cannot be met in the near-infrared and visible wavelength region. Indeed, when the fundamental wavelength falls into a broad single-resonance band (for wavelengths between about 0.4 and 2.0 $\mu$m), the $\chi_{\mathrm{QW}}^{(2)}$-tensor element $\chi_{15,\mathrm{QW}}^{(2)}$, for example, is calculated to be $\sim 10^{-10}$ m/V; this is the same order of magnitude as the $\chi_b^{(2)}$ element $\chi_{14,b}^{(2)}$ for GaAs [$3.8\times10^{-10}$ m/V (Ref. 18)]. Our AQW sample was prepared using a V80-H molecular-beam epitaxy (MBE) system. A 0.2-$\mu$m GaAs buffer layer was grown on the semi-insulating GaAs(001) substrate, which was then followed by a 0.4-$\mu$m $\mathrm{Al}_{0.8}\mathrm{Ga}_{0.2}\mathrm{As}$ buffer layer. Above it 11 periods of "opposite AQW pairs" were deposited, containing 22 single AQW's. This structure is illustrated in Fig. 1. The whole structure was terminated by a 5-nm GaAs cap layer. All of the layers were undoped. The linear refractive indices and absorption coefficients at the fundamental (775 nm) and SH (388 nm) wavelengths are obtained from Ref. 19. The absorption depth of the SH light dominates the absorption process and is approximately 13 nm in GaAs and 57 nm in $\mathrm{Al}_{0.8}\mathrm{Ga}_{0.2}\mathrm{As}$, respectively. GaAs and $\mathrm{Al}_{0.8}\mathrm{Ga}_{0.2}\mathrm{As}$ buffer layers were used to effectively eliminate SHG contribution from the substrate. With a simple model (neglecting effects such as internal reflection), we estimate that $L_b^{\mathrm{eff}}$ and $L_{\mathrm{QW}}^{\mathrm{eff}}$ are comparable (10–20 nm).

The experimental approach has been discussed in detail in Ref. 17. The source of the fundamental beam was a mode-locked Ti:sapphire laser operating at a 775-nm wavelength, generating a train of 110-fs pulses at 76 MHz with an average power of 0.8 W. The beam was focused on the sample to a 40-$\mu$m-diameter spot at an incident angle of 45°. The SHG intensity $I_{g,h}^{(2\omega)}$ was measured from a GaAs(001) wafer and an AQW sample in the $(p,s)$, $(p,p)$, and $(s,p)$ polarization configurations with $\psi$ being varied over 360° in 2° increments; the radius of the path of the beam spot on the sample surface was $\sim 1$ mm. The results for $(p,p)$ and $(s,p)$ are shown in Fig. 2. By fitting the experimental data, we deduced the relative values of the Fourier coefficients in Eq. (2),[17] as shown in Table I. The normalized $c_{g,h}^{(4)}$ are found to be $\lesssim 10^{-2}$ in all three polarization configurations. We infer that the quadrupole contribution is indeed negligible and conclude that $c_{g,h}^{(0)}$ can reasonably be attributed solely to interfacial dipole effects. Accurate retrieval of the relative phases between $c_{g,p}^{(0)}$ and $c_{g,p}^{(2)}$ proved to be difficult due to the large difference in values of $|c_{g,p}^{(0)}|$ and $|c_{g,p}^{(2)}|$. We deduced that the relative phases of the $c_{g,p}^{(0)}$ and the $c_{g,p}^{(2)}$ were within ±10° of either 0° or 180°, indicating that to a good ap-

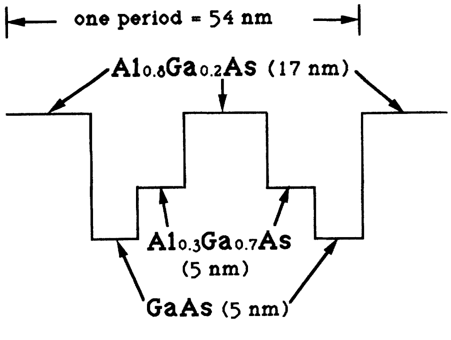

FIG. 1. Structure of a single period of the "opposite AQW pair." Only the conduction energy band is illustrated.

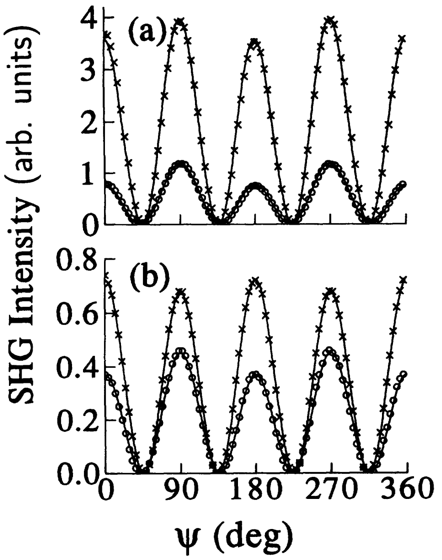

FIG. 2. SHG intensities $I_{g,h}^{(2\omega)}(\psi)$ for (a) $(p,p)$, (b) $(s,p)$. In both cases the crosses and the circles correspond to SHG from GaAs and the AQW samples, respectively. The solid lines are fits using Eq. (2a).

TABLE I. Fourier coefficients in arbitrary units deduced using Eq. (2) from SHG data obtained for GaAs and AQW samples. The error in $c_{g,h}^{(0)}/c_{g,h}^{(2)}$ is typically ±0.005. $c_{g,h}^{(4)}$ are found to be $\lesssim 10^{-2}$ for all data sets and are therefore omitted from the table.

| Polarization and Fig. | Sample type | Isotropic $c_{g,h}^{(0)}$ | Twofold $c_{g,h}^{(2)}$ | Ratio $c_{g,h}^{(0)}/c_{g,h}^{(2)}$ |
|---|---|---|---|---|
| $(p,s)$ | GaAs | $\lesssim 10^{-3}$ | 1.994 | $\lesssim 10^{-3}$ |
|  | AQW | $\lesssim 10^{-3}$ | 1.264 | $\lesssim 10^{-3}$ |
| $(p,p)$ | GaAs | −0.048 | 1.941 | −0.025 |
| Fig. 2(a) | AQW | −0.106 | 0.982 | −0.108 |
| $(s,p)$ | GaAs | 0.014 | 0.836 | 0.017 |
| Fig. 2(b) | AQW | −0.032 | 0.639 | −0.051 |

proximation the $c_{g,p}^{(0)}/c_{g,p}^{(2)}$ can be taken as real numbers.

We first consider the case of $I_{p,s}^{(2\omega)}(\psi)$. $c_{p,s}^{(2)}$ contains unseparable contributions from $C_{2v}$-type symmetry (originating from both $\chi_s^{(2)}$ and $\chi_{\mathrm{QW}}^{(2)}$, denoted in combination as $\partial_{ij}$) and $\bar{4}3m$-type (bulk) effects, respectively.[16,17] The $C_{2v}$-type contribution under $(p,s)$ comes solely from $(\partial_{15}-\partial_{24})$. $I_{p,s}^{(2\omega)}(\psi)$ from both GaAs and AQW samples was observed to display a $\sin^2(2\psi)$ behavior, but the intensity yielded by the AQW is significantly lower than that from the GaAs wafer: $c_{p,s}^{(2)}$ obtained from the AQW drops to about 0.63 times of that obtained from the GaAs. The $(p,p)$ data offer more information, since the isotropic coefficient $c_{p,p}^{(0)}$ now contains the interfacial contributions $(\partial_{31}+\partial_{32})$, $(\partial_{15}+\partial_{24})$, and $\partial_{33}$. On the other hand, $c_{p,p}^{(2)}$ contains $(\partial_{31}-\partial_{32})$ and $(\partial_{15}-\partial_{24})$, as well as the bulk $\chi_{14}^{(2)}$. These contributions cannot be resolved in this experiment. The ratio $c_{g,p}^{(0)}/c_{g,p}^{(2)}$ is a meaningful measure of the strength of the isotropic interfacial contribution relative to the bulk contribution. Yamada and Kimura[15] found this ratio to be −0.013 for and −0.037 for 2×1 and 4×6 reconstructed GaAs(001) clean surfaces, respectively, using a 580-nm fundamental wavelength under $(p,p)$. We also observed a similar effect under $(p,p)$ for both the GaAs and AQW samples. The result $c_{p,p}^{(0)}/c_{p,p}^{(2)}=-0.025$ is obtained for the GaAs sample (Table I), which is comparable to that obtained by Yamada and Kimura. However, this ratio rises sharply to −0.108 for the AQW sample, indicating a much stronger relative isotropic contribution. Meanwhile, $c_{p,p}^{(2)}$ drops to ~0.5 of that of the GaAs spectrum, the biggest relative decrease among all three polarization configurations. In the case of the $(s,p)$ configuration, the isotropic term $c_{s,p}^{(0)}$ contains only $(\partial_{31}+\partial_{32})$ and was not observed by Yamada and Kimura.[15] They therefore concluded that $\partial_{31}$ and $\partial_{32}$ are weak compared to the other $C_{2v}$-symmetry-related tensor elements, while the bulk dipole contribution still dominates in the case of pure GaAs. However, we did observe a clear $c_{s,p}^{(0)}$ in the SHG data from both the GaAs and the AQW samples (Table I). For GaAs, $c_{s,p}^{(0)}/c_{s,p}^{(2)}$ is found to be +0.017. When the AQW is examined in this polarization configuration, this ratio rises sharply in magnitude to −0.051, while $c_{s,p}^{(2)}$ reduces to 0.76 of that of GaAs. The fact that the $c_{s,p}^{(0)}$ is significant also indicates that in our case $(\partial_{15}+\partial_{24})$ and $\partial_{33}$ are not the only contributors to $c_{p,p}^{(0)}$: because $\partial_{31}$ and $\partial_{32}$ are the sole contributors to $c_{s,p}^{(0)}$, they therefore presumably contribute significantly to $c_{p,p}^{(0)}$ as well. This is very different from the results obtained by Yamada and Kimura. The difference is probably due to the different wavelength and surface preparation in the case of the GaAs(001) face, and also interfacial effects in the case of the AQW.

On the other hand, the change of $c_{g,h}^{(2)}$ observed in the AQW sample is comparable in magnitude to the $\chi_b^{(2)}$ contribution in GaAs. This is accompanied by sharp increases in $c_{g,p}^{(0)}/c_{g,p}^{(2)}$ for the AQW sample. These effects are probably not due to a change in surface reconstruction, since surface reconstruction was not observed to affect $c_{g,p}^{(0)}/c_{g,p}^{(2)}$ and the values of $c_{g,h}^{(2)}$ to this extent, albeit at a different fundamental wavelength.[15] Therefore we suggest that the sharply increased interfacial SH contribution displayed through the $c_{g,p}^{(0)}/c_{g,p}^{(2)}$ ratios mainly originates from the AQW structure. However, we should note that there are still several possible mechanisms that can contribute to the change of $c_{g,h}^{(2)}$. These mechanisms include the presence of $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ layers (with a different $\chi_{14}^{(2)}$ from that of GaAs), structural disorder in the QW interfacial region, and finally a significant but antiphased SH contribution (relative to $\chi_b^{(2)}$) from the AQW.

There are some significant disagreements in the literature concerning the value of $\chi_{14}^{(2)}$ in $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ compounds. Most reports suggested that the $\chi_{14}^{(2)}$ of $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ is either larger than or at least comparable to that of pure GaAs.[12,20] Lue, Lo, and Tzeng[12] observed that the SHG intensity obtained in reflection from MBE-grown $\mathrm{Al}_{0.3}\mathrm{Ga}_{0.7}\mathrm{As}$ bulk material is almost double that from GaAs and hence the $\chi_{14}^{(2)}$ of $\mathrm{Al}_{0.3}\mathrm{Ga}_{0.7}\mathrm{As}$ was calculated to be ~35% higher than that of GaAs. Using a different approach, Ogasawara *et al.*[20] estimated the magnitude of the nonlinear susceptibility of an $Al_xGa_{1-x}As$ waveguide to be $(1.2\pm0.5)\times10^{-10}$ m/V, which is comparable to the $\chi_{14}^{(2)}$ of GaAs. Both experiments were carried out using a 1.06-$\mu$m fundamental wavelength, falling into the same single-resonance band with the wavelength used here.[10] Bearing in mind these observations, the decrease of $c_{g,h}^{(2)}$ observed from our AQW sample is unlikely to be due to a lower SH susceptibility of the $Al_xGa_{1-x}As$ layers. On the other hand, there is at least one paper[21] claiming that the $\chi_{14}^{(2)}$ of AlAs was found to be only 0.23 times that of GaAs. Clearly, more reliable data on the values of $\chi^{(2)}$ in $Al_xGa_{1-x}As$ compounds are required. Disorder and defects, on the other hand, tend to diminish the noncentrosymmetry of the crystal, thereby reducing the magnitude of $\chi_b^{(2)}$. This effect may become significant if the defect density is high and if the volume of the disordered region is large compared to the whole volume of the SHG active region. However, previous research[12] has suggested that the interfaces of MBE-grown $Al_xGa_{1-x}As$/GaAs symmetric QW's do not significantly affect the value of $\chi_{14}^{(2)}$, indicating that good lattice order is probably preserved at these interfaces. By assuming a ~12-Å transition region for each interface, we estimate the relative interfacial volume in our AQW sample to be ~13%. The low scatter in the SHG data obtained from the AQW sample indicates excellent crystalline order on a length scale $\gtrsim 10$ $\mu$m. The large values of $c_{g,p}^{(0)}/c_{g,p}^{(2)}$ are also a sign of good interfacial order. Our tentative conclusion is that the nonlinear susceptibility of $Al_xGa_{1-x}As$ compounds is similar to that of GaAs, and that interfacial disorder plays a negligible role in the SHG properties of the AQW sample. Therefore, although at this stage we cannot completely corroborate the theoretical prediction that the magnitude of $\chi_{QW}^{(2)}$ is comparable with that of $\chi_b^{(2)}$, our experiment does show strong support for this theory. The fact that, for example, $c_{p,p}^{(0)}/c_{p,p}^{(2)}$ is negative suggests that, if the surface and AQW contributions to $c_{p,p}^{(0)}$ and $c_{p,p}^{(2)}$ are in phase, the AQW and the bulk contributions to $c_{p,p}^{(2)}$ will be antiphased, reducing the total $c_{p,p}^{(2)}$ and hence the $I_{p,p}^{(2\omega)}$ from AQW sample, as observed. In light of the confirmation of anticipated enhancement from a single AQW, it is now clear that QPM structures based on the AQW's have the potential for significant overall enhancement in SHG with appropriate choice of subband gap (visible and/or near-infrared) excitation wavelength. We are currently carrying out such experiments on a set of QPM AQW samples at 1.06-$\mu$m fundamental wavelength, and expect to report these results in the near future. Approaches for further investigation also include modifying the phase difference between the SH fields generated from the AQW's and the bulk, possibly by changing the growth sequence and depth from the surface of the AQW's.

It is also worth mentioning that previously SH spectroscopy has been shown to be sensitive to interfacial traps, lattice relaxation, and buried reconstruction of semiconductors.[22] Our observation of strong interfacially sensitive isotropic terms in SHG data from noncentrosymmetric compounds using short-wavelength light further demonstrates the applicability of reflected SHG as a powerful probe of buried heterojunction structures in these compounds.

In conclusion, we have observed a clear isotropic contribution in SHG rotational data obtained from both a $GaAs/Al_xGa_{1-x}As$ AQW sample and a GaAs(001) wafer for $p$-polarized SH radiation. We believe that this is also the first report of an observation of such a contribution in noncentrosymmetric materials in the $(s,p)$ configuration. Evidence is obtained that the $\chi^{(2)}$ of the AQW is similar to that of GaAs, in agreement with theoretical work. The azimuthal dependence of the SHG intensity is shown to be useful in investigating both surfaces and heterostructural interfaces in a wide range of noncentrosymmetric compound materials.

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