APPLIED PHYSICS LETTERS **90**, 151915 (2007)

Magnetic-field-induced fourfold azimuthal angle dependence in the terahertz radiation power of (100) InAs

Elmer Estacio,[a] Hisashi Sumikura, Hidetoshi Murakami, Masahiko Tani, Nobuhiko Sarukura, and Masanori Hangyo
Institute of Laser Engineering, Osaka University, 2-6 Yamadaoka, Suita, Osaka 565-0871, Japan

Carlito Ponseca, Jr., Romeric Pobre,[b] and Reuben Quiroga[b]
Institute for Molecular Science, 38 Nishigonaka, Myodaiji, Okazaki 444-8585, Japan

Shingo Ono
Nagoya Institute of Technology, Gokiso, Showa, Nagoya, Aichi 466-8555, Japan

(Received 8 December 2006; accepted 9 March 2007; published online 12 April 2007)

The azimuthal angle dependence in the terahertz radiation power of (100) InAs under 1 T magnetic field is presented. Results show that although the dominant radiation mechanism is surge current, azimuthal-angle-dependent radiation due to the nonlinear effect is also observed. The twofold symmetry of the *p*-polarized terahertz radiation power was modified to a fourfold symmetry with the transverse magnetic field. Moreover, results exhibited fourfold symmetry for the *s*-polarized terahertz power even with no applied field. The anisotropic intervalley scattering of photocarriers is tentatively proposed as the origin of quadrupole response and the fourfold emission symmetry. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2721385]

The generation of intense terahertz radiation from InAs surfaces illuminated with ultrashort optical pulses has been the subject of immense work in the past decade.[1,2] Moreover, the behavior of this radiation phenomenon for various applied magnetic field strengths and excitation fluence has been thoroughly investigated.[3–8] The continued research efforts on the terahertz radiation mechanisms in surface-illuminated InAs wafers, however, imply that a complete understanding of the terahertz radiation phenomena still has to be realized. Recently, several works on the influences of carrier concentration, diffusion, drift, and excitation wavelength dependence on InAs terahertz radiation have been reported. This lends proof to the contention that much has yet to be done in this area of research.[2,9–11]

In this letter, we present experimental work on the azimuthal angle dependence of the polarized terahertz radiation power of a (100) *p*-type InAs wafer. It has been revealed that the dominant terahertz radiation mechanism in our study is due to an azimuthal-angle-independent mechanism. This angularly independent contribution could originate from photocarrier diffusion, surge current, and/or electric-field-induced optical rectification.[6,12,13] Moreover, further investigation on the dependence of the terahertz time domain spectroscopy (TDS) wave form on the polarity of the applied magnetic field supports the dominance of the surge current mechanism. However, an angle-dependent, twofold, nonlinear bulk optical rectification effect was also detected. In the presence of an external 1 T magnetic field applied parallel to the sample surface, the twofold terahertz emission symmetry was modified to a fourfold symmetry for the *p*-polarized terahertz radiation. This fourfold dependence was also observed for the *s*-polarized terahertz radiation even with no applied field. An explanation pertaining to the anisotropic *X* or *L* valley scatterings is offered to account for the results. Knowledge of such carrier scattering is important in the design considerations for InAs-based terahertz emitters.

van der Pauw–Hall measurements were firstly performed to determine the carrier concentration and the carrier mobility of the *p*-type InAs (100) sample at room temperature. The measured carrier concentration was $7.1\times10^{16}\ \mathrm{cm}^{-3}$ and the hole mobility was $150\ \mathrm{cm}^2/\mathrm{V\,s}$. Primarily, terahertz TDS and excitation fluence dependence of the terahertz radiation power were measured to obtain some information on the mechanism of terahertz radiation. The TDS experiments were performed in free space with an excitation wavelength of 800 nm from a *p*-polarized, mode-locked Ti: sapphire laser with a 100 fs pulse width and a repetition rate of 82 MHz. The average pump power was maintained at 160 mW and the beam diameter at the sample was ~0.5 mm. The angle of incidence of the laser was set at 45° with respect to the sample surface normal, and the terahertz radiation was collected in the reflection direction using suitable optics. The transverse applied field was provided by a 1 T permanent magnet. Steady-state power measurements were also performed to investigate the excitation fluence and azimuthal angle dependence of the terahertz radiation power as described by Gu and Tani.[6] The azimuthal angle dependence was measured for *p*-and *s*-polarized terahertz radiation components. In this case, the photoconductive antenna was replaced by an InSb hot-electron bolometer.

In order to discriminate the magnetic-field-enhanced terahertz radiation, the TDS wave forms for the B_{up}, B_{down} ($=-B_{\mathrm{up}}$), and no-field cases were taken (illustrated in the inset of Fig. 1). This offers an insight on the dominant terahertz radiation mechanism as previously reported.[5,14] Shown in Fig. 1 are the wave forms obtained by subtracting the TDS wave form of the no-field case from the B_{up} and B_{down} data. It is evident from the figure that the wave forms were π phase shifted with respect to each other. Migita and Hangyo have shown that the Lorentz-force-driven terahertz radiation

---

[a] Author to whom correspondence should be addressed; electronic mail: estacio-es@ile.osaka-u.ac.jp

[b] Permanent address: Department of Physics, De La Salle University, Taft Avenue, Manila 1004, Philippines.

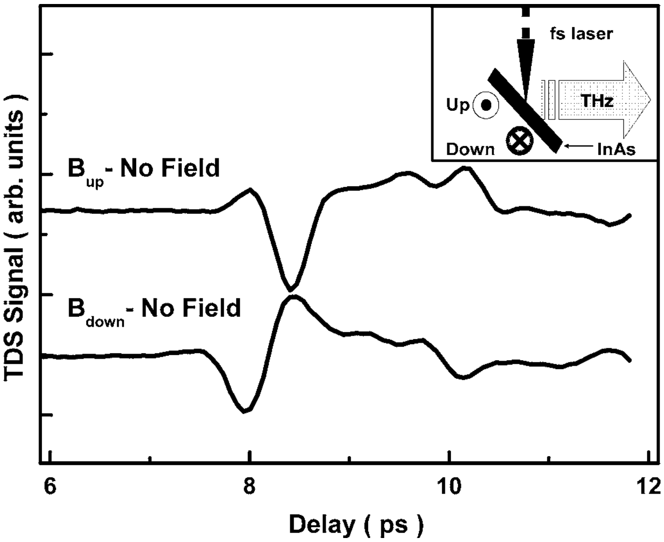

FIG. 1. Subtracted TDS wave forms for the $B_{\mathrm{up}}-$no field and $B_{\mathrm{down}}-$no field cases where the orientation of the applied magnetic field is shown in the inset. The π-shifted wave forms clearly show the reversal of the dominant Lorentz-force-driven current surge when the field direction was flipped.

due to the drift current will have a π-phase-shifted TDS wave form when the applied magnetic field direction is flipped due to the reversal of the drift current direction.[14] Moreover, excitation fluence dependence experiments revealed that up to a fluence of 1.2 mJ/cm^2, the dominant terahertz radiation mechanism is due to photocarrier diffusion as shown in Fig. 2. This is in agreement with a previous report demonstrating that, for excitation fluences below 2 mJ/cm^2, an angle-independent terahertz radiation mechanism is dominant.[15] It is worthwhile to emphasize that the TDS data and the subsequent terahertz power azimuthal angle dependence were all measured with an excitation fluence of $\sim$0.2 mJ/cm^2, well below the saturation of surge current terahertz radiation.

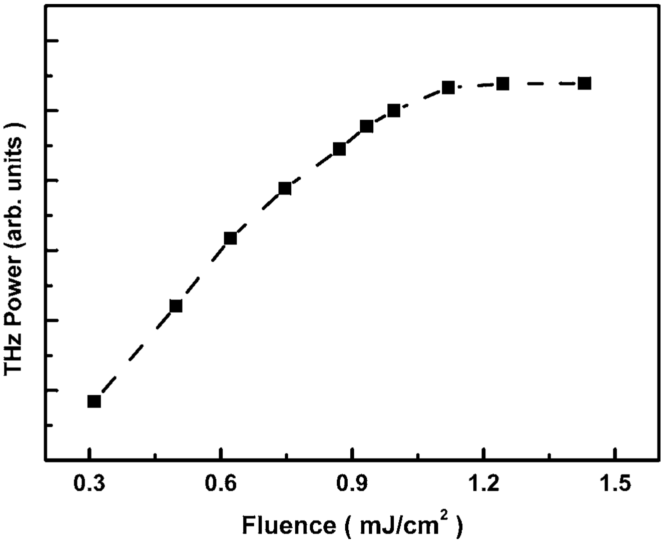

FIG. 2. Excitation fluence dependence of the emitted terahertz radiation. The photocarrier-related radiation mechanism saturates at about 1.2 mJ/cm^2. All the subsequent azimuthal-angle-dependence measurements were performed at an excitation fluence of 0.2 mJ/cm^2, wherein the surge current terahertz radiation is expected to be dominant.

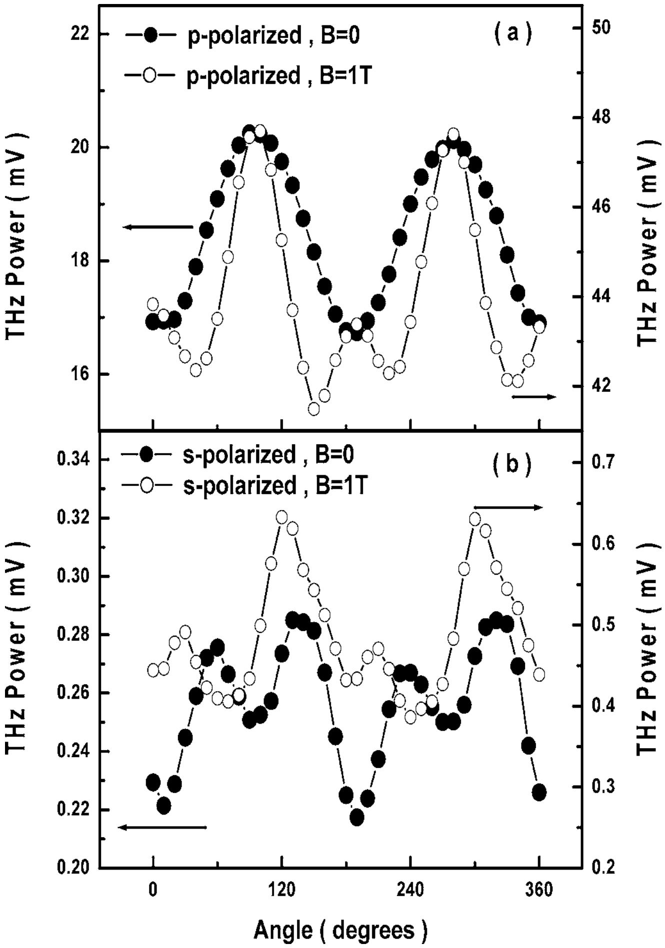

FIG. 3. (a) Azimuthal angle dependence of the p-polarized terahertz radiation power (the voltage values are the actual lock-in amplifier readouts). The twofold symmetry of the bulk optical rectification effect for (100) InAs was modified to a fourfold symmetry with the transverse magnetic field. (b) Weak fourfold dependence is observed in the s-polarized terahertz radiation even with no applied field. These results are suggested to be due to the photocarrier scattering to the X or L valleys.

The azimuthal angle dependence plots of the terahertz radiation power are shown in Fig. 3. The y axes are the voltage readouts from the lock-in amplifier-detected bolometer output, wherein the noise levels did not exceed 20 μV. The azimuthal angle, on the other hand, is defined as the angle between the crystallographic axis ($[\bar{1}10]$ or $[1\bar{1}0]$) and the reflection plane. A twofold azimuthal symmetry for the p-polarized radiation with no applied field was observed as shown by the closed circle trace in Fig. 3(a). The oscillatory signal is superposed on a high dc offset and constitutes only about 20% of the total terahertz radiation power. Evidently, although terahertz radiation from an angularly dependent mechanism is clearly observed, the dominant terahertz radiation mechanism is due to an angle-independent surge current phenomenon, either from the surface depletion field or from the photo-Dember effect.[6] The open circle trace shows the behavior of the with-field case. The magnetic-field-induced enhancement due to the increased radiation collection efficiency is immediately seen.[16,17] The power was observed to increase by about 140%. Interestingly, a fourfold symmetric, azimuthal-angle-dependent feature appeared to be superposed on the usual twofold dependence. Initially, this modified symmetry was surmised to originate from a nonlinear effect due to a magnetization-induced nonlinear optical susceptibility χ^{magn} in InAs. The magnetization-induced $\chi^{(2)}$ contribution is given by[18]

$$P^{\mathrm{nl}}(\Omega \sim 0 = \omega - \omega) = \chi^{\mathrm{cr}}E(\omega)E(-\omega) + \chi^{\mathrm{magn}}E(\omega)E(-\omega)M = 2\varepsilon_0 \sum_{jkl} \chi^{(2)}_{ijkl}E_jE_kH_l. \tag{1}$$

However, a calculation of the magnetization-induced nonlinear susceptibility tensor in Eq. (1), taking into account the $\bar{4}3m$ and (100) orientation for InAs, failed to yield the fourfold symmetry that was observed in the experiment. The possible modification from a twofold to a fourfold azimuthal symmetry has been initially suggested by Reid and Fedosejevs to be attributed to a quadrupole response of InAs. This, however, has been previously expected to have insignificant contribution.[6,15]

The azimuthal angle dependence of the much weaker *s*-polarized terahertz radiation power is shown in Fig. 3(b). Even with no applied field, the distinct fourfold symmetry is evident even as the dc offset still dominates the terahertz radiation. With an applied magnetic field, the dc offset also increased and the fourfold symmetry shifted slightly but was not modified. It appears that the fourfold azimuthal dependence is a weak contribution, easily drowned by the much more intense *p*-polarized, twofold symmetric, bulk-optical-rectification-induced terahertz radiation. Previous reports on the azimuthal angle dependence of *s*-polarized terahertz radiation have not demonstrated this fourfold symmetry, presumably due to inherently weak signal levels and reduced signal-to-noise ratio. Moreover, an incomplete separation of the two polarized components will not manifest as the distinct symmetry folding as these have been previously reported to be pahse shifted by just ~45°.[15]

This weak fourfold symmetric behavior that was enhanced by the applied magnetic field (in the *p*-polarized terahertz radiation case) is possibly attributed to a weak quadrupole response from an anisotropic intervalley scattering to the L and X valleys in the $\langle 111\rangle$ and $\langle 110\rangle$ directions, respectively. With ~1.55 eV optical excitation and considering that the holes will also have excess energy, the photogenerated electrons have an excess energy of ~1.03 eV for InAs (E_G =0.35 eV). This is sufficient for the photocarriers to be scattered to these two high-symmetry satellite valleys from the Γ valley ($E_X - E_G = 1.02$ eV and $E_L - E_G = 0.73$ eV).[19] A fourfold azimuthal angle dependence was previously reported in the terahertz emission of yttrium barium copper oxide thin films via bulk electric quadrupole–magnetic dipole optical rectification.[20] Although, this terahertz radiation mechanism has been previously reported to be insignificant in InAs, it is deduced that the intervalley scatterings of carriers in four equivalent directions in the crystallographic half space imply the creation of an electric quadrupole moment both in the absence and presence of the applied field. This quadrupole response is thought to cause the fourfold symmetry regardless of the actual terahertz radiation mechanism being surge current or a nonlinear optical effect. With a transversely applied field, the quadrupole- and dipole-related emissions may be enhanced with the associated tilting of their electric moments from the sample surface normal according to the Lorentz force.[17] As the wafer is rotated about its surface normal, the Lorentz-force-driven carriers will move in either the positive or negative Y directions in the laboratory frame (i.e., parallel to the sample surface), depending on the field polarity.[6–17] Thus, at several azimuthal orientations, the direction of the motion of the carriers will coincide with the $\langle 110\rangle$ (or $\langle \bar{1}\bar{1}0\rangle$) directions or the projection of the $\langle 111\rangle$ (or $\langle \bar{1}\bar{1}1\rangle$) directions to the InAs surface (which is equivalent to the $\langle 110\rangle$ or $\langle \bar{1}10\rangle$ directions).

The intervalley scattering time does not exceed 100 fs and is much shorter than the estimated cyclotron period of ~800 fs under a 1 T applied field for electrons in InAs ($m_e^* = 0.023m_o$).[21,22] Although the acceleration of the carriers due to the cyclotron motion itself is the origin of the enhanced terahertz radiation under an applied field, the modification of the carrier trajectory can be neglected in the context of valley scattering.

In summary, the terahertz radiation from (100) *p*-InAs was studied, revealing dominant radiation from the surge current for an excitation fluence of ~0.2 mJ/cm^2. The twofold azimuthal dependence of the *p*-polarized terahertz radiation power due to bulk optical rectification was modified to a fourfold symmetry under 1 T magnetic field. This was also exhibited by the *s*-polarized terahertz radiation power even with no applied field. An anisotropic intervalley scattering mechanism in four equivalent directions that gives rise to a quadrupole response is tentatively proposed for this observation.

The authors thank K. M. A. Bautista and A. Salvador for technical assistance. This research is supported by the Grant-in-Aid for Creative Scientific Research from the Japan Society for the Promotion of Science. Two of the authors (R.P. and R.Q.) are visiting as Associate Professors at the Institute of Molecular Science.

[1] N. Sarukura, H. Ohtake, S. Izumida, and Z. Liu, J. Appl. Phys. **84**, 654 (1998).

[2] R. Adomavicius, G. Molis, A. Krotkus, and V. Sirutkaitis, Appl. Phys. Lett. **87**, 261101 (2005).

[3] M. B. Johnston, D. M. Whitaker, A. Corchia, A. G. Davies, and E. H. Linfield, Phys. Rev. B **65**, 165301 (2002).

[4] Matthew Reid and Robert Fedosejevs, Appl. Opt. **44**, 149 (2005).

[5] C. Weiss, R. Wallenstein, and R. Beigang, Appl. Phys. Lett. **77**, 4160 (2000).

[6] P. Gu and M. Tani, in *Terahertz Optoelectronics*, Topics in Applied Physics Vol. 97, edited by K. Sakai (Springer, Berlin, 2005), pp. 63–76.

[7] H. Ohtake, S. Ono, M. Sakai, Z. Liu, T. Tsukamoto, and N. Sarukura, Appl. Phys. Lett. **76**, 1398 (2000).

[8] R. H. Takahashi, A. Quema, R. Yoshioka, S. Ono, and N. Sarukura, Appl. Phys. Lett. **83**, 1068 (2003).

[9] K. Liu, J. Xu, T. Yuan, and X.-C. Zhang, Phys. Rev. B **73**, 155330 (2006).

[10] G. Chern, E. Readinger, H. Shen, M. Wraback, C. Gallinat, G. Koblmuller, and J. Speck, Appl. Phys. Lett. **89**, 141115 (2006).

[11] M. Suzuki, M. Tonouchi, K. Fujii, H. Ohtake, and T. Hirosumi, Appl. Phys. Lett. **89**, 091111 (2006).

[12] M. Reid, I. V. Cravetchi, and R. Fedosejevs, Phys. Rev. B **72**, 035201 (2005).

[13] R. Adomavičius, A. Urbanowicz, G. Molis, A. Krotkus, and E. Šatkovskis, Appl. Phys. Lett. **85**, 2463 (2004).

[14] M. Migita and M. Hangyo, Appl. Phys. Lett. **79**, 3437 (2001).

[15] M. Reid and R. Fedosejevs, Appl. Phys. Lett. **86**, 011906 (2005).

[16] J. Shan, C. Weiss, R. Wallenstein, R. Beigang, and T. F. Heinz, Opt. Lett. **26**, 849 (2001).

[17] M. B. Johnston, D. M. Whittaker, A. Corchia, A. G. Davies, and E. H. Linfield, J. Appl. Phys. **91**, 2104 (2002).

[18] R. R. Birss, *Symmetry and Magnetism* (North-Holland, Amsterdam, 1966), pp. 62–66.

[19] S. Adachi, J. Appl. Phys. **66**, 6030 (1989).

[20] S. J. L. W. Siders, S. A. Trugman, F. H. Garzon, R. J. Houlton, and A. J. Taylor, Phys. Rev. B **61**, 2013634 (2000).

[21] P. Y. Yu and M. Cardona, *Fundamentals of Semiconductors: Physics and Materials Properties*, 2nd ed. (Springer, Berlin, 1999), p. 216.

[22] R. Ascazubi, I. Wilke, K. J. Kim, and P. Dutta, Phys. Rev. B **74**, 075323 (2006).