Excitation fluence dependence of terahertz radiation mechanism from femtosecond-laser-irradiated InAs under magnetic field

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The excitation fluence and magnetic field dependence of terahertz (THz) radiation power from InAs is investigated. At low excitation fluence, an enhancement of the THz-radiation power is observed independent of the magnetic-field direction. As the excitation fluence is increased, a crossover of the terahertz radiation mechanism is observed. At excitation fluence above this crossover, the radiation power is either enhanced or reduced depending on the magnetic-field direction. These results are explained by considering the different THz-radiation mechanisms from the InAs surface with or without photoexcited carrier screening.
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Since the frequency region of terahertz (THz) radiation is between the well-developed microwave with gigahertz frequency and laser with PHz frequency, practical scheme to generate intense THz radiation has not been established until recently. Up to now, many frontier works are still considered to be part of this region. Utilizing ultrafast optical pulses, various THz-radiation emitters including photoconductive switches,[1] semiconductor surface,[2] and nonlinear optical process[3] have been reported. Due to the remarkable progress of these devices, many applications, such as sensing/imaging and time-resolved spectroscopy, were successfully demonstrated in the far-infrared region.[4,5] To facilitate these applications, a compact and coherent light source is strongly required. In view of these, semiconductor surfaces have attracted a great deal of attention. After Zhang *et al.* reported the enhancement of THz-radiation power from GaAs under an external magnetic field,[6] various semiconductors have been used as THz-radiation emitters.[7–10] Recently, InAs irradiated by near-infrared optical pulses is recognized as a viable emitter for real-world applications.[11–14] This is because of its narrow band gap and smaller effective mass than that of GaAs. The origin of THz radiation from the semiconductor surface is generally categorized into two mechanisms, namely, acceleration of photoexcited carriers by the surface depletion-field and current-surge induced by the different diffusion velocities between photoexcited electrons and holes. However, the mechanism of THz radiation from InAs has not been clarified completely.[15–18] This is partly due to the lack of experimental results providing clear insights to explore it.

In this letter, we report the dependence on optical excitation fluence of THz-radiation power from InAs using a Ti:sapphire regenerative amplifier laser system. Such a system enables us to investigate the effect of excitation fluence over a wide range. It is found that the magnetic-field dependence of THz-radiation power exhibits completely different behavior depending on the excitation fluence. For low excitation fluence, the THz-radiation power increases regardless of the magnetic field direction. In contrast, for high excitation fluence, the THz-radiation power increases or decreases depending on the magnetic field direction. These results can be explained by taking into account the different mechanisms of THz-radiation from InAs.

The experimental setup for the THz-radiation emitter is shown in Fig. 1. A Ti:sapphire regenerative amplifier laser system, which delivered 100 fs optical pulses at a center wavelength of 800 nm, is used as an excitation source. The laser provided 1 W average power with a repetition rate of 5 kHz. A 75° angle of incidence, which is near the Brewster angle, was selected and the laser spot area on the sample surface was approximately 2.4 cm^2.[19] An undoped, slightly *n*-type InAs bulk crystal with (100) surface and carrier density (N) of 5×10^{16} cm^{-3} was used as the sample. By using an electromagnet, the magnetic field was applied parallel to the sample surface with maximum field strength of about 1.7 T. THz-radiation power was detected by a liquid-helium-cooled Ge bolometer (QGEB/2F:QMC Instrument Ltd.), and

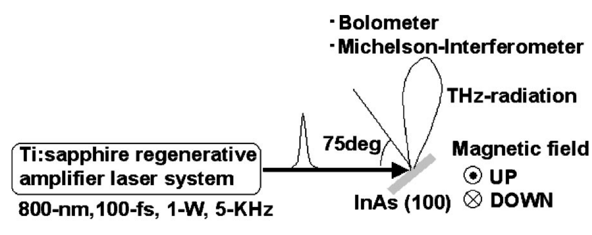

FIG. 1. Experimental setup for the THz radiation in a magnetic field. A Ti:sapphire regenerative amplifier laser system is used as an excitation source. The incident angle of 75° was selected, which is near the Brewster angle. The sample used was an undoped, slightly *n*-type InAs bulk crystal with (100) surface. THz-radiation power was detected by a liquid-helium-cooled Ge bolometer (QGEB/2F:QMC Instruments Ltd), and the THz-radiation spectrum was obtained by a Michelson interferometer.

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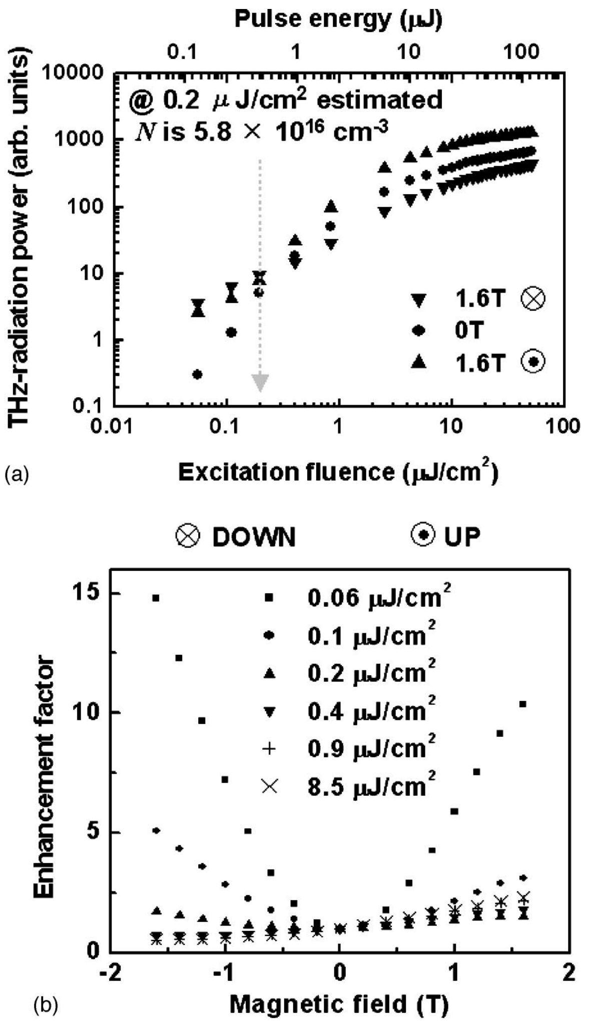

FIG. 2. (a) The effect of excitation fluence on the magnetic-field induced enhancement of THz-radiation power. The dotted arrow points to the excitation fluence of 0.2 μJ/cm^2, which corresponds to a photoexcited carrier density of 5.8×10^{16} cm^{-3}. (b) Magnetic-field dependence of THz-radiation power from an excitation fluence of 0.06–0.9 μJ/cm^2.

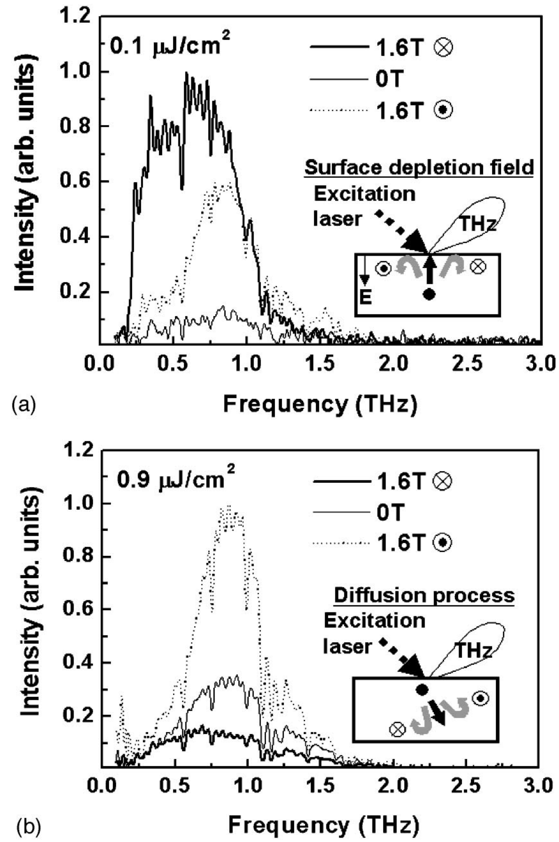

FIG. 3. THz-radiation spectra measured at excitation fluences of (a) 0.1 and (b) 0.9 μJ/cm^2. The insets show the schematic diagrams of electron motion under (a) surface depletion field and (b) diffusion process. Arrows show the electron motion with (gray) and without magnetic field (black).

the THz-radiation spectra were obtained by a Michelson interferometer.

Figure 2(a) presents the effect of excitation fluence on the magnetic-field induced enhancement of THz-radiation power. All results consistently shows that THz-radiation average power increases with excitation fluence and exhibits saturation at higher excitation fluence. Such saturation can be attributed to the large amount of electron-hole pairs generated via optical absorption.[20] The generation of electron-hole pairs increases the carrier–carrier scattering rate. This causes the THz radiation to be less coherent leading to a less efficient THz-radiation emission. A crossover of the THz-radiation mechanism is also observed at around 0.2 μJ/cm^2 excitation fluence. This crossover is further discussed in Fig. 2(b). The magnetic-field induced enhancement factor of THz-radiation power at various excitation fluences is shown in Fig. 2(b). At low excitation fluence below 0.2 μJ/cm^2, the THz-radiation power is enhanced and exhibits quadratic dependence for both directions of the magnetic field as observed in the previous report.[13] Additionally, the enhancement factor of the THz-radiation power is observed to be higher for the down direction as compared to the up direction. However, increasing the excitation fluence drastically changes this situation. At excitation fluence of around 0.2 μJ/cm^2, the deviation from the quadratic magnetic-field dependence is clearly observed, and the enhancement factors for both magnetic-field directions are dramatically reduced. With increasing excitation fluence over 0.2 μJ/cm^2, the THz-radiation power increases or decreases depending on the magnetic-field direction. The magnetic-field dependence of THz-radiation power at excitation fluence over 8.5 μJ/cm^2 overlapped that of 0.9 μJ/cm^2. For the up direction, THz-radiation power increases with magnetic field at much smaller enhancement factor than that of the low excitation fluence. Furthermore, for the down direction, THz-radiation monotonically decreases with magnetic field up to about 1.6 T. The radiation spectra were obtained using a Michelson interferometer to further investigate these phenomena. Figure 3 presents the THz-radiation spectra at excitation fluences 0.1 and 0.9 μJ/cm^2. With an excitation fluence of 0.1 μJ/cm^2, the spectral wave form of THz radiation changes significantly depending on magnetic-field direction. A shift of the spectral weight towards the low frequency region is observed for the down direction. In contrast, for the 0.9 μJ/cm^2 excitation fluence no such shift is observed and the spectral wave form of THz radiation is unchanged.

To explain these phenomena, we consider the two mechanisms for THz radiation from the InAs surface, i.e., the acceleration under the surface depletion field and the diffusion process. THz radiation could also be generated via the optical rectification, which is described by the second-order nonlinear susceptibility [$\chi^{(2)}$]. However, for InAs(100), the contribution of this process in generating THz radiation is observed to be much smaller (6%) than that of the surge current as reported by Gu *et al.*[8] The reason of this is that the (100) surface has a symmetric structure, while the nonzero $\chi^{(2)}$ results from the broken symmetry induced by the surface depletion field and off-normal incidence of the excitation laser.[21] Moreover, unlike the previously reported $\chi^{(2)}$ originated THz radiation from InAs (111),[22] the excitation fluence in our experiment is two order smaller even at higher excitation case. Therfore, surge current should be the dominant mechanism for the generation of THz radiation. Schematic diagrams of the carrier motion are depicted as insets in Fig. 3. By considering an excitation fluence of 0.2 μJ/cm^2, the estimated photoexcited carrier density is 5.8×10^{16} cm^{-3} from the penetration depth of 140 nm at an optical excitation of 800 nm.[23] This value is comparable to the doping density of the sample, which is approximately 5×10^{16} cm^{-3}. At low excitation fluence, the photoexcited carrier density is below 5×10^{16} cm^{-3} and the screening effect of the surface depletion field is still not significant. In this case, the photoexcited carriers are accelerated by the surface depletion field and acquire a drift velocity along the surface normal as shown in the inset of Fig. 3(a). The Lorentz force, which is a cross product between this drift velocity and the magnetic field, changes the direction of the carrier acceleration toward the surface parallel. Due to the dielectric contrast between air and semiconductor surface, this change of dipole direction significantly enhances the THz radiation. The difference of THz-radiation spectrum caused by magnetic-field direction is due to the reversal of acceleration direction, which inverts the phase of THz radiation as confirmed by THz time-domain spectroscopy.[22] On the other hand, at high excitation fluence, the surface depletion field is almost screened out and the diffusion process becomes significant. Diffusion process is initiated by the carrier-density gradient, and its direction is given by the incident angle of excitation pulses, not normal to the surface as shown in the inset of Fig. 3(b). By applying a magnetic field, the dipole is rotated to the direction in which the THz radiation is effectively or ineffectively extracted from the surface depending on the magnetic-field direction.[18] Thus, the spectral wave form of the THz radiation remains unchanged with magnetic-field direction, and only the radiation power is affected. It should be pointed out that the diffusion velocity is proportional to the carrier-density gradient, which is given by the absorption coefficient of InAs, and not affected by the excitation fluence. Since the dipole rotation is induced by the Lorentz force, which is a cross product of the diffusion velocity and the magnetic field, the magnetic-field dependence of THz-radiation power overlaps at higher excitation fluence. The reduction of the enhancement factor at higher excitation fluence might be attributed to the carrier–carrier scattering effect, which plays a more important role rather than the Lorentz force.

In summary, the dependence on optical-excitation fluence of THz-radiation power from InAs is investigated using Ti:sapphire regenerative amplifier laser system as an excitation source. THz-radiation power exhibits completely different behavior depending on the excitation fluence. These results are explained by taking into consideration the different mechanisms of THz radiation from InAs. At low excitation fluence, the THz radiation mainly originates from the carrier acceleration by the surface depletion field and the THz-radiation power is enhanced regardless of magnetic field direction. In contrast, at high excitation fluence, the surface depletion field is almost screened out and the diffusion process becomes significant. This process becomes the main mechanism for THz radiation and the radiation power is either enhanced or reduced depending on the magnetic-field direction.

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