Research Article | Vol. 30, No. 7 / 28 Mar 2022 / Optics Express 11789

Optics EXPRESS

Increase in terahertz-wave intensity in a magnetic field due to difference-frequency mixing by exciton excitation in a GaAs/AlAs multiple quantum well

OSAMU KOJIMA,[1,*] YUKI TARUI,[1] TAKASHI KITA,[1] AVAN MAJEED,[2] PAVLO IVANOV,[2,3] EDMUND CLARKE,[2] AND RICHARD A. HOGG[2,3]

[1] *Department of Electrical and Electronic Engineering, Graduate School of Engineering, Kobe University, 1-1 Rokkodai, Nada, Kobe 657-8501, Japan*
[2] *Department of Electronic and Electrical Engineering, University of Sheffield, Sheffield S3 7HQ, United Kingdom*
[3] *Electronics and Nanoscale Engineering, James Watt School of Engineering, University of Glasgow, Glasgow G12 8LT, United Kingdom*
[*] *kojima@phoenix.kobe-u.ac.jp*

Abstract: Magnetic fields can increase the intensity of terahertz (THz) waves due to changing the dipole moment direction using the Lorentz force. This study reports the increase in the THz-wave intensity generated by differential frequency mixing using commercial permanent magnets under exciton-excitation. While a weak magnetic field applied to a multiple quantum well increases the THz-wave intensity due to excitons, a strong field causes its decrease. According to the calculations, the increase is caused by the electron-hole separation due to the Lorentz force. Furthermore, the calculations suggest the importance of carrier acceleration to enhance the intensity. Importantly, the increase in the THz-wave intensity due to differential frequency mixing does not require a strong magnetic field and can be achieved with inexpensive commercially available magnets.

1. Introduction

Difference-frequency mixing (DFM) is an effective method to produce monochromatic terahertz (THz) waves with a certain frequency in a wide frequency range. It has attracted much attention, because THz waves with a narrow linewidth are required to detect specific molecules with unique absorption frequencies in e.g. human breath or the atmosphere [1–5]. Previously, we reported a wide frequency tunability of THz waves by DFM using excitons in multiple quantum wells (MQWs) [6,7]. Herein, while the exciton optical nonlinearity enhances the THz-wave intensity around the optical transition energy of a heavy-hole (HH) exciton confined in an MQW, no enhancement was observed at the simultaneous excitation of HH and light-hole (LH) excitons. Furthermore, the overlap of the exciton states, for instance, the HH and LH excitons in different MQWs, can increase the DFM signal [8], but that requires a complicated sample structure.

An internal electric field plays an important role in breaking the centrosymmetry of the envelope functions in the process of THz-wave generation by DFM in semiconductors. Usually, as is well known, semiconductor crystals have a surface electric field caused by surface states [9,10], hence, the electron-hole dipole is aligned along the growth direction, as shown in Fig. 1(a). In our samples, the MQWs were embedded in a pin structure where excitons are longitudinally polarized, creating the second nonlinear polarization due to breaking the centrosymmetry. On the contrary, the direction of laser light polarization is usually parallel to the surface, decreasing the coupling efficiency between light and dipoles. Therefore, changing the dipole direction is one

#453812 https://doi.org/10.1364/OE.453812
Journal © 2022 Received 14 Jan 2022; revised 14 Mar 2022; accepted 15 Mar 2022; published 23 Mar 2022

Optics EXPRESS

of the methods to enhance the THz signal as shown in Fig. 1(b). Indeed, applying a magnetic field enhances the THz signal generated by ultrashort pulses [11–16]. Meanwhile, the coupling efficiency between light and excitons should be enhanced in the in-plane direction of the exciton dipole, hence increasing the THz-wave intensity due to DFM. In the QW systems, the exciton binding energy is enhanced, so that the electron and hole may be tightly bounded. Nevertheless, in the case of GaAs/AlAs MQWs, the exciton binding energy is approximately 10 meV [17], which is less than the thermal energy at room temperature. Then, we considered that the excitons created in the MQW can be easily changed to the electron-hole pairs. In fact, previously, we reported the possibility of ultrafast dephasing of excitons in a GaAs/AlAs MQW [18]. From these points, we considered that the change in the dipole can be described with the same model. Therefore, this study deals with the effect of changing the exciton dipole direction by permanent magnets on the intensity of THz waves due to DFM, because the magnetic effects on the continuous THz wave generation is very important for aplications such as the THz spectroscopy. The excitation-power and excitation-energy dependences show an increase in the THz-wave intensity by the magnetic field. Furthermore, the results of our calculations indicate that a strong magnetic field is not required to increase the signal intensity. The change in the THz-wave intensity is discussed on the basis of the dependence of the exciton dipole alignment.

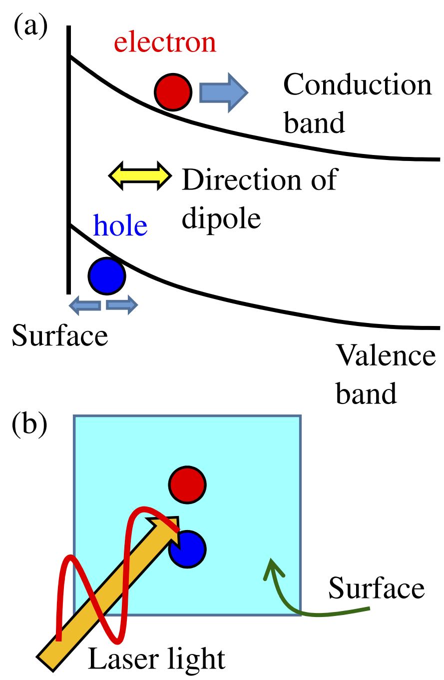

Fig. 1. (a) Direction of carrier movement. The electron precedes the hole because of the surface field. (b) Ideal carrier configuration to enhance exciton coupling with light.

2. Experiment

We used an undoped GaAs/AlAs MQW embedded in a pin structure on a (001) n^+-doped GaAs substrate grown by molecular-beam epitaxy. Detailed information on the sample, including the exciton energies, and the measurement system configuration is available in our previous report [6]. The MQW consists of 30 alternating layers of GaAs and AlAs with a thickness of 7.35 nm each. The parameters of terahertz waves were measured at 297 K. A semiconductor laser with a photon energy of 1.506 eV, and a continuous-wave mode Ti:sapphire laser were used to vary

Optics EXPRESS

the excitation energy. The two beams were combined on a half mirror in free space with the same polarization, and the combined beam was focused on the sample surface. The terahertz signal was detected by a pyroelectric sensor (THZ2I-BL-BNC, Gentech-EO) and amplified by a lock-in-amplifier at 5 Hz. To apply the magnetic field, commercial neodymium magnets manufactured by SIMOTECH Co., Ltd. [19] were attached to the sample, as shown in Fig. 2(a). A nominal magnetic field ranging from 0.2 to 0.5 T was applied by changing the magnets. The magnetic-field direction was parallel to the surface, while an iron plate was installed on the opposite side to suppress field spread. Because of the p-i-n structure, electrons created by the laser light moved toward the substrate. Therefore, the Lorentz force worked along the z-direction as shown in Fig. 2(b). The polarization component in the plane is created as the hole current direction is opposite to that of electrons.

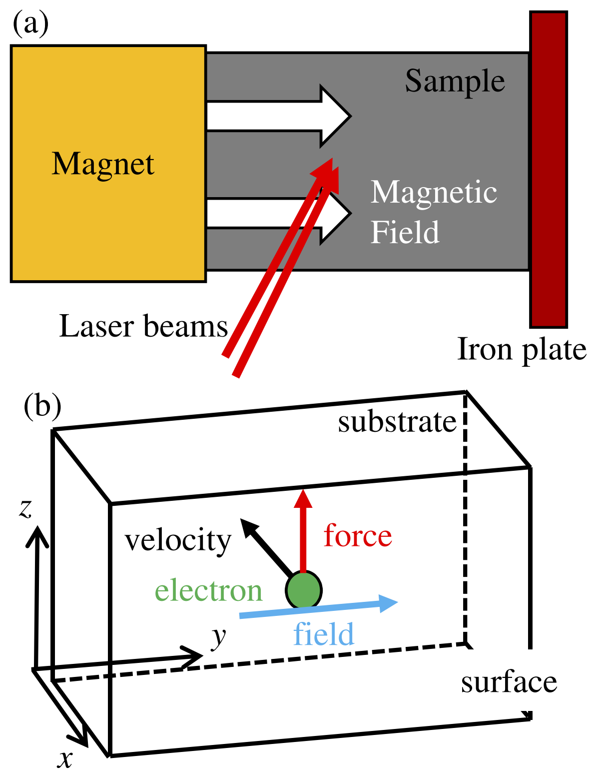

Fig. 2. (a) Configuration of the magnet, sample, and iron plate. (b) Schematic of the directions of the magnetic field, Lorentz force, and electron velocity directions. It is noted that the direction of the current is opposite to the velocity.

3. Results and discussion

Figure 3 demonstrates the signal intensities measured as a function of excitation power under the magnetic fields of 0.3 and 0.4 T. In these measurements, the lasers with the intensities ratio of 1:5 excited HH and LH excitons, respectively [6]. The intensity demonstrates the square dependence on the excitation power, which is a characteristic of the second-order optical nonlinear effects causing the emission of THz wave due to the DFM process [6,7]. because the amplitude of the terahertz wave is given by the product of those of two excitation sources. Furthermore, the intensity at 0.4 T is larger than that at 0.3 T due to the stronger magnetic field.

Optics EXPRESS

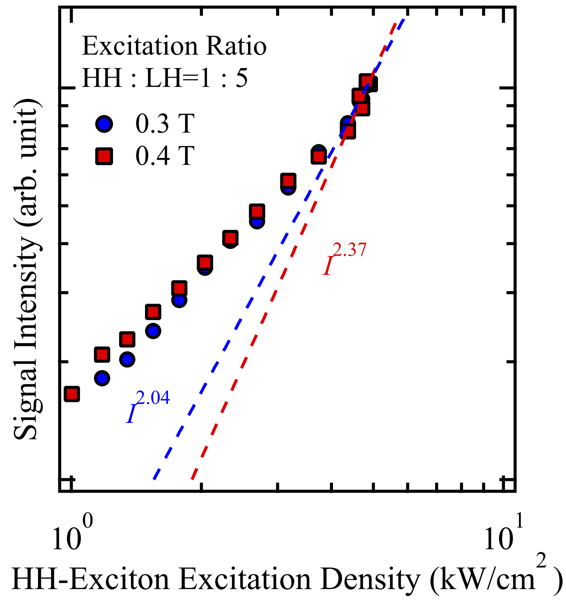

Fig. 3. Signal intensity measured at various excitation power under the magnetic fields of 0.3 and 0.4 T indicated by the circles and squares, respectively. The dashed lines are least-square fits.

Next, the magnetic-field effect is studied at different excitation energies, i.e., the energy of the Ti:sapphire laser (Fig. 4). The magnetic field of up to 0.4 T basically enhanced the signal intensity. In particular, at the LH exciton energies, the intensity ratio depends on the magnetic field . This increase is caused by the change in the exciton dipole alignment from the MQW growth direction to the in-plane direction.

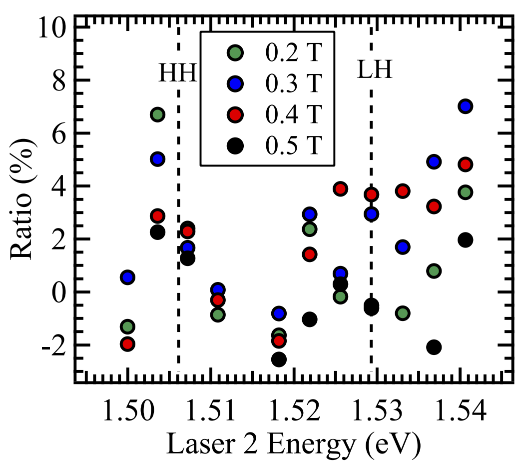

Fig. 4. Increment of the signal intensity (the intensity ratio to that without magnetic field) at different field strength and various excitation energies (energy of Ti:sapphire laser). The dotted lines indicate the heavy-hole (HH) and light-hole (LH) exciton energies [6].

On the contrary, the intensity rapidly decreases at 0.5 T; in particular, at the HH exciton energy, the intensity is maximal at 0.2 T and gradually decreases with the magnetic field. In the case of the LH-exciton energy, the maximum is at 0.4 T. To discuss the dependence of the signal intensity on the magnetic field strength, the carrier displacement caused by the Lorentz force was calculated. Here, the previous explanations related to the enhancement of the terahertz waves by magnetic fields have been developed under pulse excitation conditions. On the other hand, the basic idea related to the change in the direction of the dipole and carrier displacements is a Hall effect which is based on the continuous current. Then, the results measured with the continuous lasers are discussed on the basis of the Hall effect. When the electron and hole velocities become larger, the larger dipole will be generated because the electrons and holes can move longer during their lifetime. According to Fig. 2(b), the electron current, magnetic field, and the Lorentz force are aligned along the x, y, z axes, respectively. Based on the explanation of the Hall effect, the electron velocities v_{ex}, v_{ey}, and v_{ez} are described as follows:

$$v_{ex} = -\frac{e\tau E_x}{m_e} + \frac{e\tau v_{ez} B}{m_e} \tag{1}$$

$$v_{ey} = 0 \tag{2}$$

$$v_{ez} = -\frac{e\tau v_{ex} B}{m_e}, \tag{3}$$

where e is the elementary charge, E_x is applied electric field strength, τ is lifetime, m_e is electron effective mass, and B is magnetic field strength (strictly speaking, magnetic flux density). Moreover, it was assumed that the Lorentz force does not move carriers in the x and y directions. In the usual Hall effect, E_z corresponds to the Hall electric field, and v_{ez} vanishes because the sample cross-section is fulfilled by the carriers. On the other hand, in this measurement, the carriers are created within the laser spot; the carriers disappear outside of the laser spot. According to this, E_z vanishes and v_{ez} remains. By substitution of (3) into (1),

$$v_{ex} = -\frac{e\tau E_x m_e}{m_e^2 + e^2\tau^2 B^2} \tag{4}$$

$$v_{ez} = \frac{e^2\tau^2 E_x B}{m_e^2 + e^2\tau^2 B^2} \tag{5}$$

are derived. Using the value of $E_x = 20.6$ kV cm^{-1} for the built-in field caused by the p-n junction estimated from the doping concentrations, the dependencies of v_{ex} and v_{ez} on the magnetic field were calculated with various lifetimes from 50 fs to 2 ps as shown in Figs. 5(a) and 5(b), respectively. The electron effective mass of $0.066m_0$ [20], where m_0 is free electron mass, was used in the calculation. The negative sign of the velocity indicates that electrons move toward the substrate. The velocities for HH and LH, v_{Hx}, v_{Hz}, v_{Lx}, and v_{Lz}, were also calculated [Figs. 5(c)–5(f)] using the effective masses m_H and m_L of $0.34m_0$ and $0.097m_0$ [20], respectively, and the Eqs. (6) and (7):

$$v_{H(L)x} = \frac{e\tau E_x m_{H(L)}}{m_{H(L)}^2 + e^2\tau^2 B^2} \tag{6}$$

$$v_{H(L)z} = \frac{e^2\tau^2 E_x B}{m_{H(L)}^2 + e^2\tau^2 B^2}. \tag{7}$$

Here, Eqs. (5) and (7) are the same sign, which results from $E_z = 0$. This is different from the equations of usual Hall effect.

The velocities in the x-direction, i.e., the growth direction, decrease due to the applied magnetic field. On the other hand, the velocities in the z-direction caused by the Lorentz force demonstrate a different behavior: while the dependences of electron and LH velocities possess a clear maxima;

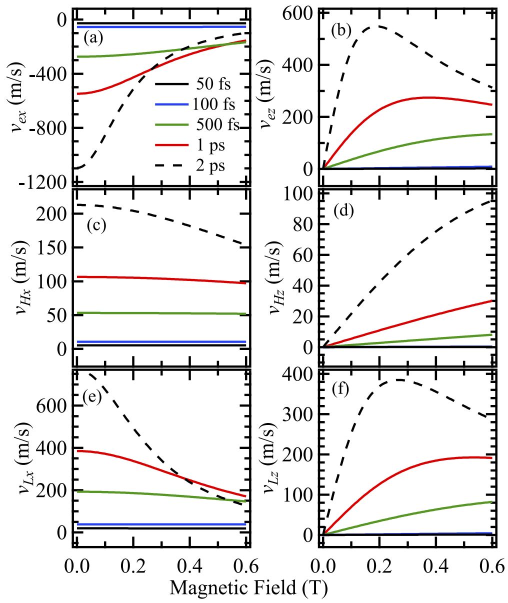

Fig. 5. Calculated velocities of electrons with various lifetimes as a function of the magnetic field in the x- (a) and z-(b) directions. For holes, the velocities of heavy-hole (HH) and light-hole (LH) are plotted for the x- and z-directions in (c)-(f), respectively.

v_{Hz} simply increases with the magnetic field. This difference is attributed to the effective masses: the heavier the mass, the stronger the magnetic field at the peak velocity.

The change in the separation length of electrons and holes caused by the magnetic field was calculated assuming that the electron and hole lifetimes are the same according to the continuous-wave excitation. Using the values of each velocity, $\Delta_{eh} = \sqrt{(v_{ex}\tau - v_{H(L)x}\tau)^2 - (v_{ez}\tau - v_{H(L)z}\tau)^2}$ were calculated. The large separation may induces the exciton ionization, which reduces the signal intensity. The results for HH and LH excitons are shown by the solid and dotted curves in Fig. 6(a), respectively. Because the typical Δ_{eh} is below 1 nm, the decrease in the overlap integral corresponding to the oscillator strengths hardly affects the signal intensity. This suggests that the quantum-confined Stark effect has more impact on electron-hole spatial separation than the Lorentz force.

In addition, separation in the z-direction Δ_z was calculated, as shown in Fig. 6(b). Interestingly, the value of Δ_z depends on the magnetic field. In particular, the peak appears in the weaker fields at longer lifetimes.

Returning to results in Fig. 4, the intensity at the HH exciton energy was maximal at 0.2 T. Only HH excitons were excited in that condition. Therefore, the dependence of Δ_z, which can be considered as the coupling strength projection, on the magnetic field strongly affects the signal intensity. On the contrary, the intensity reached the maximum at 0.4 T in the case of excitation of both HH and LH excitons. This result is difficult to explain. However, we assume that v_{Lz} is maximal when the coupling of HH and laser light is the strongest around 0.2 T. Namely, the signal may be enhanced not only due to the exciton-light coupling but also by carrier acceleration. Moreover, the intensity in the linear increase region in Fig. 3 is also changed. This is also attributed to the change in the velocity of z-direction because the current density depends on the velocity.

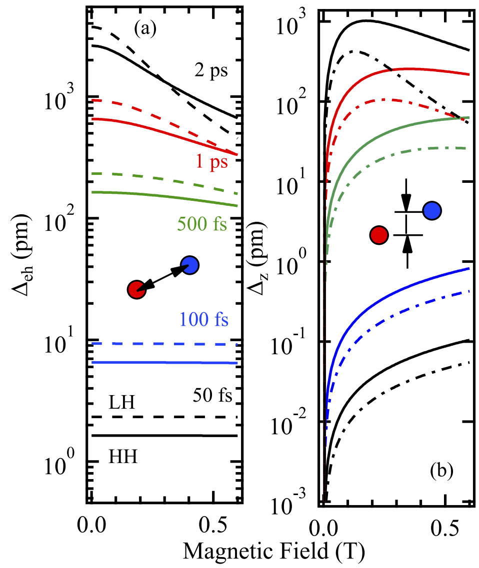

Fig. 6. (a) Calculated spatial separation between electron and hole Δ_{eh} caused by the Lorentz force for various lifetime values. The solid and dotted curves indicate the results for electron-heavy-hole (HH) and electron-light-hole (LH), respectively. (b) z-direction component of the spatial separation.

4. Conclusion

The magnetic-field effects on the DFM signal intensity in the THz range were studied under exciton-excitation conditions as a function of excitation intensity and energy. The intensity of the THz wave was enhanced by the magnetic field at weaker field strengths, whereas it decreased under a stronger field. According to the calculations, carrier lifetimes are strongly related to the intensity enhancement factor. In particular, a stronger magnetic field was not required around the HH exciton energy. These results indicate that the proposed simple and cost effective method enables the THz signal enhancement by using weak commercial magnets.

Funding. Japan Society for the Promotion of Science (16KK0129, 19K04532, 21K03435, 26289088); JSPS International Research Fellow; Kawanishi Memorial ShinMaywa Education Foundation; Support Center for Advanced Telecommunications Technology Research Foundation; MIKIYA Science and Technology Foundation; Hyogo Foundation for Science and Technology.

Acknowledgments. The author would like to thank Enago for the English language review.

Disclosures. The authors declare no conflicts of interest.

Data availability. Data underlying the results presented in this paper are not publicly available at this time but may be obtained from the authors upon reasonable request.

References

1. K. A. McIntosh, E. R. Brown, K. B. Nichols, O. B. McMahon, W. F. DiNatale, and T. M. Lyszczarz, "Terahertz photomixing with diode lasers in low-temperature-grown GaAs," Appl. Phys. Lett. **67**(26), 3844–3846 (1995).
2. M. A. Belkin, F. Capasso, A. Belyanin, D. L. Sivco, A. Y. Cho, D. C. Oakley, C. J. Vineis, and G. W. Turner, "Terahertz quantum-cascade-laser source based on intracavity difference-frequency generation," Nat. Photonics **1**(5), 288–292 (2007).
3. T. Kruczek, R. Leyman, D. Carnegie, N. Bazieva, G. Erbert, S. Schulz, C. Reardon, S. Reynolds, and E. U. Rafailov, "Continuous wave terahertz radiation from an InAs/GaAs quantum-dot photomixer device," Appl. Phys. Lett. **101**(8), 081114 (2012).
4. Q. Y. Lu, S. Slivken, N. Bandyopadhyay, Y. Bai, and M. Razeghi, "Widely tunable room temperature semiconductor terahertz source," Appl. Phys. Lett. **105**(20), 201102 (2014).

Optics EXPRESS

5. B. Liu, H. Bromberger, A. Cartella, T. Gebert, M. Först, and A. Cavalleri, "Generation of narrowband, high-intensity, carrier-envelope phase-stable pulses tunable between 4 and 18 THz," Opt. Lett. **42**(1), 129 (2017).
6. O. Kojima, Y. Tarui, H. Shimazu, T. Kita, A. Majeed, P. Ivanov, E. Clarke, and R. Hogg, "Wide frequency tuning of continuous terahertz wave generated by difference frequency mixing under exciton-excitation conditions in a GaAs/AlAs multiple quantum well," Phys. Rev. Appl. **10**(4), 044035 (2018).
7. A. Majeed, P. Ivanov, B. Stevens, E. Clarke, I. Butler, D. Childs, O. Kojima, and R. Hogg, "Broadband THz absorption spectrometer based on excitonic nonlinear optical effects," Light: Sci. Appl. **8**(1), 29 (2019).
8. K. Sakaue, O. Kojima, T. Kita, M. J. Steer, and R. A. Hogg, "Increase in terahertz-wave generation by difference frequency mixing by overlap of exciton states in different GaAs/AlAs quantum wells and spectroscopic measurements," Opt. Express **29**(15), 24387 (2021).
9. J. L. Shay, "Photoreflectance Line Shape at the Fundamental Edge in Ultrapure GaAs," Phys. Rev. B **2**(4), 803–807 (1970).
10. D.E. Aspnes, "Third-derivative modulation spectroscopy with low-field electroreflectance," Surf. Sci. **37**, 418–442 (1973).
11. R. McLaughlin, A. Corchia, and M. B. Johnston, "Enhanced coherent terahertz emission from indium arsenide in the presence of a magnetic field," Appl. Phys. Lett. **76**(15), 2038–2040 (2000).
12. C. Weiss, R. Wallenstein, and R. Beigang, "Magnetic-field-enhanced generation of terahertz radiation in semiconductor surfaces," Appl. Phys. Lett. **77**(25), 4160–4162 (2000).
13. J. N. Heyman, P. Neocleous, D. Hebert, P. A. Crowell, T. Müller, and K. Unterrainer, "Terahertz emission from GaAs and InAs in a magnetic field," Phys. Rev. B **64**(8), 085202 (2001).
14. A. Corchia, R. McLaughlin, M. B. Johnston, D. M. Whittaker, D. D. Arnone, E. H. Linfield, A. G. Davies, and M. Pepper, "Effects of magnetic field and optical fluence on terahertz emission in gallium arsenide," Phys. Rev. B **64**(20), 205204 (2001).
15. M. Hangyo, M. Migita, and K. Nakayama, "Magnetic field and temperature dependence of terahertz radiation from InAs surfaces excited by femtosecond laser pulses," J. Appl. Phys. **90**(7), 3409–3412 (2001).
16. M. B. Johnston, D. M. Whittaker, A. Corchia, A. G. Davies, and E. H. Linfield, "Theory of magnetic-field enhancement of surface-field terahertz emission," J. Appl. Phys. **91**(4), 2104–2106 (2002).
17. R. L. Greene, K. K. Bajaj, and D. E. Phelps, "Energy levels of Wannier excitons in GaAs-$Ga_{1-x}Al_xAs$ quantum-well structures," Phys. Rev. B **29**(4), 1807–1812 (1984).
18. O. Kojima, T. Kita, M. J. Steer, and R. A. Hogg, "Resonant exciton excitation photoluminescence and dynamics in a GaAs/AlAs multiple quantum well with internal electric field," AIP Adv. **10**(9), 095016 (2020).
19. Company website for the magnets (Japanese): https://www.simotec.co.jp/products/magnet/mg02/mg02-01/.
20. O. Madelung, *Semiconductors: Data Handbook* (Splinger, Berlin, 2003).