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| **Citation** | Journal of Applied Physics, 110(1): 013515 |
| **Issue Date** | 2011-07-07 |
| **Type** | Journal Article |
| **Textversion** | publisher |
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| **DOI** | 10.1063/1.3603046 |





TAKEUCHI H., TSURUTA S., & NAKAYAMA M. (2011). Terahertz spectroscopy of dynamics of coupling between the coherent longitudinal optical phonon and plasmon in the surge current of instantaneously photogenerated carriers flowing through the i-GaAs layer of an i-GaAs/n-GaAs epitaxial structure. *Journal of Applied Physics*. 110.

# Terahertz spectroscopy of dynamics of coupling between the coherent longitudinal optical phonon and plasmon in the surge current of instantaneously photogenerated carriers flowing through the *i*-GaAs layer of an *i*-GaAs/*n*-GaAs epitaxial structure

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(Received 4 April 2011; accepted 22 May 2011; published online 7 July 2011)

We demonstrate the dynamics of coupling between the coherent longitudinal optical (LO) phonon and plasmon of instantaneously photogenerated electrons in an undoped GaAs/*n*-type GaAs (*i*-GaAs/*n*-GaAs) epitaxial structure using time-domain terahertz spectroscopy. Initially, we experimentally and numerically clarify the presence of the built-in electric field in the *i*-GaAs layer of the *i*-GaAs/*n*-GaAs epitaxial layer. Next, we performed the terahertz-wave measurements of the *i*-GaAs/*n*-GaAs epitaxial structure at various excitation conditions from a low density excitation regime to a high excitation regime. The LO-phonon-plasmon coupled (LOPC) mode has been confirmed from the terahertz-wave measurement. It is found that the frequency of the LOPC mode is determined by the pump-beam power. This fact demonstrates that the LOPC mode is formed in the *i*-GaAs layer. In addition, we performed the time-partitioning Fourier transform in order to reveal the dynamical change in the LOPC mode as a function of time delay. Using this analysis, we have observed that the disappearance of the LOPC mode immediately occurs within the time delay of 0.6 ps. Following the disappearance of the LOPC mode, only the bare coherent GaAs LO phonon dominates the terahertz waves. 
[doi:10.1063/1.3603046]

## I. INTRODUCTION

Terahertz spectroscopy[1,2] is a useful way to reveal an interesting issue in the fundamental aspect of the dynamics of instantaneously photogenerated carriers. In compound semiconductors, the terahertz electromagnetic wave is produced by the illumination of a pump beam composed of femtosecond laser pulses. The illumination of the pump beam instantaneously generates carriers and launches a surge current. This surge current causes the emission of the terahertz wave. In the research field of the terahertz wave, in general, the dynamics of the surge current is simply regarded as just a flow from the surface to the internal side of a bulk crystal.[3–8] Here, we emphasize that the surge current itself is a flow of the collection of photogenerated carriers. Consequently, the collective motion of the carriers, the so-called plasmon, should coexist in the surge current. In other words, the surge current of the photogenerated carriers also has the characteristics of the plasmon. The above-mentioned interesting viewpoint, however, has been little discussed so far. We also note that the plasmon strongly couples with a longitudinal optical (LO) phonon under a high density excitation condition. Accordingly, the surge current of the photogenerated carriers has a large amount of information on the dynamical interaction between the carriers and LO phonons, and hence the investigation of the surge current of the photogenerated carriers has a great impact on ultrafast optical science and technology. It should be noted that the dynamics of the coherent LO-phonon-plasmon coupled (LOPC) mode is significant information since the feedback of the obtained information to an epitaxial structure design is useful to develop a frequency tunable terahertz emitter.

In the present work, using time-domain terahertz spectroscopy, we have investigated the LOPC mode formed by the surge current flowing through the undoped GaAs (*i*-GaAs) layer of an *i*-GaAs/*n*-type GaAs (*n*-GaAs) epitaxial structure. The present paper is organized as follows. First, we introduce the present sample structure with use of a numerical simulation and characterize the built-in electric field in the *i*-GaAs layer using photoreflectance spectroscopy. Second, we describe the results of the terahertz-wave measurement and demonstrate the formation of the LOPC mode, the frequency of which is determined only by a pump-beam power. Third, we perform the time-partitioning Fourier transform in order to demonstrate the evidence of the disappearance of the LOPC mode. It is observed that the LOPC mode dominates in the initial stage of the time evolution of the terahertz wave within the time delay of 0.6 ps and that the only terahertz wave from the bare coherent LO phonon is followed by disappearance of the LOPC mode. Here, we emphasize that the present approach is quite different from earlier works by Kersting *et al.*[9] and by Hasselbeck *et al.*[10] The earlier works, in fact, reported on the LOPC mode in

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*n*-type InAs and GaAs crystals using terahertz spectroscopy. In these reports, it was concluded that the frequency of the LOPC mode is determined only by a carrier concentration originating from dopants. In addition, the earlier works did not treat the dynamical change in the coherent LO phonon after the coupling with the plasmon: the disappearance process. Finally, we summarize the present work.

## II. SAMPLE STRUCTURE AND CHARACTERIZATION OF THE BUILT-IN ELECTRIC FIELD

The present sample was an *i*-GaAs(200 nm)/*n*-GaAs(3 $\mu$m and $3 \times 10^{18}$ cm$^{-3}$) epitaxial structure grown on a semi-insulating (001) GaAs substrate by metal organic vapor phase epitaxy, where the values of the parentheses denote the individual layer thickness and doping concentration. Figure 1 shows the equilibrium potential structure in the *i*-GaAs/*n*-GaAs epitaxial structure calculated with use of a computational simulation on the basis of the Boltzmann-Poisson model.[11,12] The parameters used in the calculations are the same as those used in Ref. 13. It should be noted that the surface Fermi level of *i*-GaAs locates at almost the center of the bandgap.[13,14] In Fig. 1, the solid line indicates the conduction-band energy calculated as a function of distance from the surface, where the origin of the energy axis corresponds to the Fermi level (dashed line). The conduction-band energy has a finite linear potential slope in the *i*-GaAs layer induced by the surface Fermi-level pinning. The resultant phenomenon produces a uniform built-in electric field that is estimated to be 35 kV/cm. The built-in electric field accelerates photogenerated carriers and causes a surge current. In addition, the built-in electric field in the *i*-GaAs layer sweeps out the background free electrons; namely, the *i*-GaAs layer is depleted. This fact is the key point of the present sample; namely, the *i*-GaAs top layer plays a role in the emission zone of the terahertz waves that are related to the coherent GaAs LO phonons and photogenerated carriers.

Next, we measured the photoreflectance spectrum at room temperature in order to experimentally evaluate the built-in electric field. In the photoreflectance measurement,

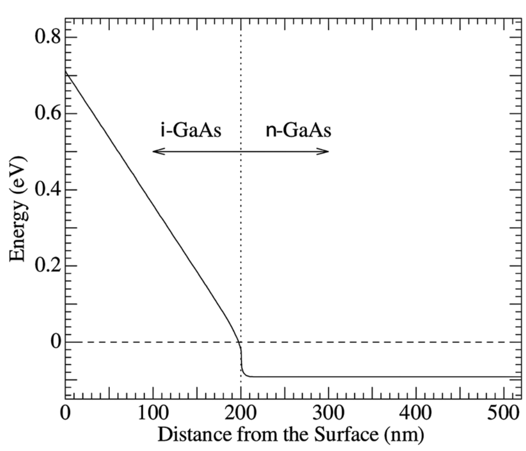


FIG. 1. Potential energy of the *i*-GaAs/*n*-GaAs epitaxial structure as a function of distance from the surface calculated on the basis of the Boltzmann-Poisson model. The solid and dashed lines indicate the conduction-band energy and Fermi level, respectively.

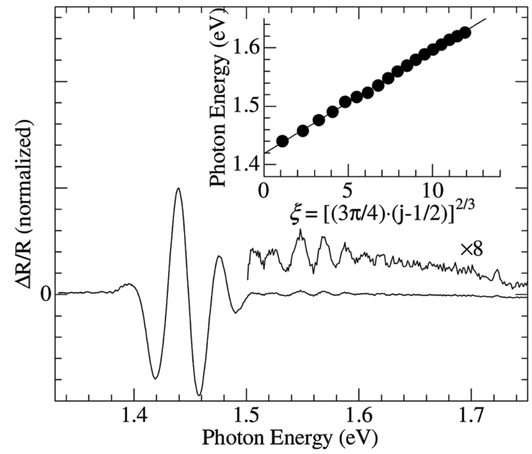


FIG. 2. Photoreflectance spectrum of the *i*-GaAs/*n*-GaAs epitaxial structure at room temperature. The inset shows the plot of the extrema of the FKOs as a function of the quasi-index $\xi$. The solid line in the inset indicates the fitted result.

the pump beam was the laser light with the photon energy of 1.96 eV chopped at a frequency of 630 Hz. The pump-beam power was about 2.0 mW. The probe beam was obtained from a tungsten-halogen lamp dispersed by a monochromator with a resolution of 0.5 nm. The probe-beam power was about 4 $\mu$W. Figure 2 shows the photoreflectance spectrum of the *i*-GaAs/*n*-GaAs epitaxial structure. In the photoreflectance spectrum, the oscillation patterns, the so-called Franz-Keldysh oscillations (FKOs), are observed. Since the FKOs result from an electric field, the appearance of the FKOs indicates the presence of the built-in electric field in the *i*-GaAs layer. In order to estimate the built-in electric field, as shown in the inset of Fig. 2, the extrema of the FKOs are plotted as a function of the quasi-index $\xi \equiv [(3\pi/4)\cdot(j-1/2)]^{2/3}$, where $j$ denotes the index of each extremum numbered from the fundamental transition energy position.[15] The slope of the solid line is the electro-optic energy $\hbar\Theta$ given by $(e^2\hbar^2F^2/2\mu)^{1/3}$, where $F$ and $\mu$ are the built-in electric field and interband reduced effective mass, respectively.[15] The built-in electric field is estimated from the observed FKOs to be 28 kV/cm using the relation of $\hbar\Theta = (e^2\hbar^2F^2/2\mu)^{1/3}$. In this estimation of the built-in electric field, the value of $\mu$ used is that of a bulk GaAs crystal: 0.0556 in units of the electron rest mass ($m_0$).[16] Thus, the *i*-GaAs layer is experimentally confirmed to be depleted. Note that the presence of the built-in electric field enables the photogenerated electrons to escape from the *i*-GaAs layer, which is discussed in detail in Sec. VI B.

## III. EXPERIMENTAL PROCEDURE FOR TIME-DOMAIN TERAHETRZ MEASUREMENT

The signals of the time-domain terahertz wave from the *i*-GaAs/*n*-GaAs epitaxial structure were measured using laser pulses with a duration time of about 60 fs. The repetition rate of the laser pulse was about 90 MHz. The pump beam was focused on the sample with the incidence angle of 45°. The diameter of the spot on the sample surface was about 120 $\mu$m. The photon energies of the pump and gate beams were the same: 1.57 eV. The absorption coefficient and refractive index for GaAs of the present pump beam are $1.4 \times 10^4$ $cm^{-1}$ and 3.69, respectively.[17] Taking account of these values, for example, in the illumination of the pump beam with 50 mW, the density of the photogenerated carriers is estimated to be $1.2 \times 10^{17}$ $cm^{-3}$. The emitted terahertz wave was collected with use of two off-axis parabolic mirrors, and was detected by an optically gated dipole antenna with a gap of 6.0 $\mu$m formed on a low-temperature-grown GaAs layer. The power of the gate beam was fixed to 10 mW. All the measurements were performed at room temperature. The humidity was controlled to be 10% during the measurement under a nitrogen-gas-purge condition. The scan range of the time delay was from − 2.0 to 8.0 ps.

## IV. EXPERIMENTAL RESULTS AND DISCUSSION

### A. Emission of the terahertz wave from the LOPC mode

Figure 3(a) shows the terahertz waveforms at various pump-beam powers in the time delay range from − 1.5 to 4.5 ps.

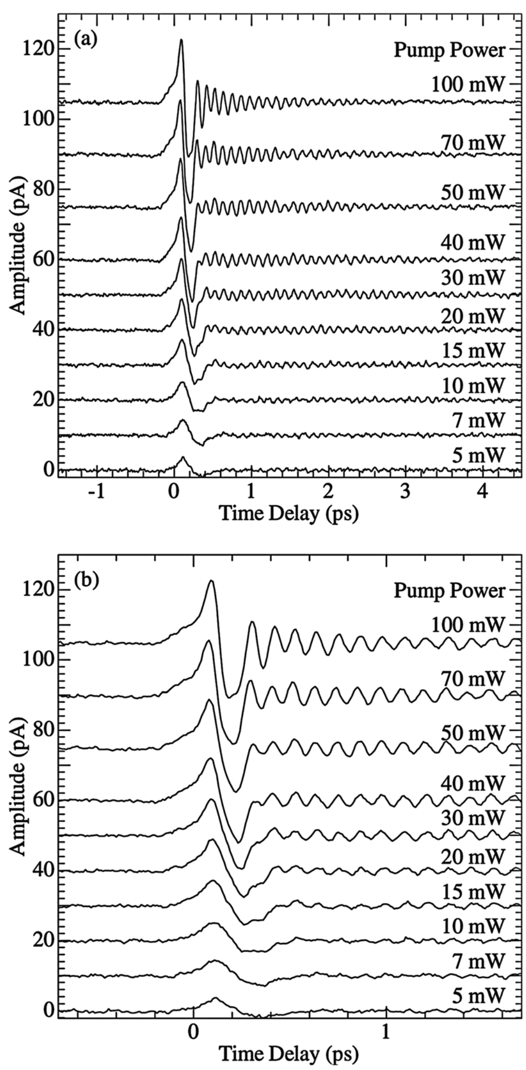


FIG. 3. (a) Amplitudes of the terahertz waveforms of the *i*-GaAs/*n*-GaAs epitaxial structure at various pump-beam powers in the time-delay range from − 1.5 to 4.5 ps. For clarity, each waveform is vertically shifted. (b) Amplitudes of the terahertz waveforms shown in Fig. 3(a) in the time-delay range from − 0.5 to 1.5 ps.

As shown in Fig. 3(a), the monocycle signals, which are usually attributed to the surge current component, are observed around the time delay of 0 ps in all the terahertz waveforms. In addition, the terahertz waveforms are followed by an oscillatory pattern with a period of about 110 fs. Here, we focus our attention on the pattern of the monocycle signal. In order to highlight the monocycle signal, the terahertz waveform in the time range between − 0.5 and 1.5 ps are extracted and depicted in Fig. 1(b). It is evident that the shape of the monocycle signal is modified with an increase in the pump-beam power.

In order to clarify the components of the terahertz waveform, we performed the Fourier transform, which is shown in Fig. 4. At first, we focus our attention on the band of the monocycle signal in the frequency range from 0 to 7 THz. The band shows an asymmetric shape with a tail extending to the high frequency side at pump-beam powers higher than 20 mW, whereas the peak frequency is fixed at about 2.0 THz. It is, therefore, considered that the tail on the higher frequency is different from the component of the surge current. In the much higher density excitation regime (70 and 100 mW), the tail on the higher frequency side exhibits a remarkable change; namely, the broad band apparently splits to two components. This fact supports the above-mentioned consideration that the tail on the higher frequency side is different from the component of the surge current. Next, we discuss the terahertz band at 8.8 THz. The peak frequency of 8.8 THz is the same as the frequency of the GaAs LO phonon. Accordingly, the band at 8.8 THz is attributed to the coherent GaAs LO phonon band. We note that the coherent GaAs LO phonon bands have an asymmetric tail on the higher frequency side at the pump-beam powers higher than 20 mW. The above-mentioned findings, (1) the split of the band originating from the monocycle and (2) the tail of the coherent GaAs LO phonon band, suggest that the formation of the coherent GaAs LOPC mode, which is pointed out in Sec. I.

In advance to decomposing the Fourier power spectrum for the estimation of the frequency of the LOPC mode, we

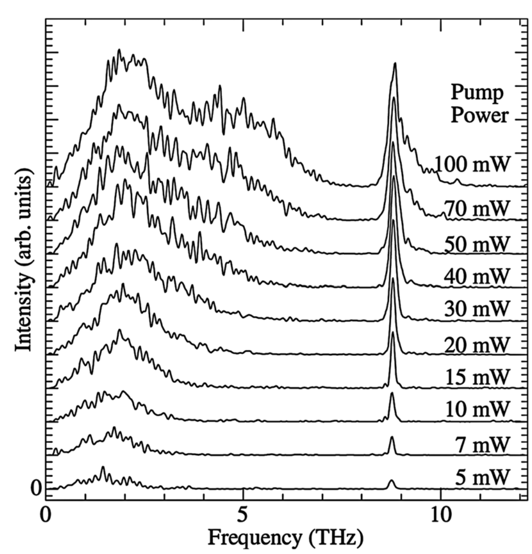


FIG. 4. Fourier power spectra of the terahertz waveforms of the *i*-GaAs/*n*-GaAs epitaxial structure at various pump-beam powers.

discuss the appropriateness of the decomposition to investigate the formation of the LOPC mode in terms of the so-called phonon strength $S_{\text{ph}}^{\text{L}+(-)}$. The phonon strength is expressed by the following equation:[18]

$$S_{ph}^{L_\pm} = \frac{\pm\omega_{L_\pm}^2 \mp \omega_p^2}{\omega_{L_+}^2 - \omega_{L-}^2}. \quad (1)$$

Here, $\omega_{\text{L}+(\text{L}-)}$ and $\omega_{\text{p}}$ are the frequency of the upper (lower) branch of the LOPC mode and the frequency of the plasmon, respectively, which are given by[19]

$$\omega_{L\pm}^2 = \frac{1}{2}\left[\left(\omega_{\text{p}}^2 + \omega_{\text{LO}}^2\right) \pm \sqrt{\left(\omega_{\text{p}}^2 + \omega_{\text{LO}}^2\right)^2 - 4\omega_{\text{p}}^2\omega_{\text{TO}}^2}\right] \quad (2a)$$

and

$$\omega_{\text{p}} = \sqrt{\frac{ne^2}{\varepsilon_\infty m^*}}. \quad (2b)$$

In Eq. (2a), $\omega_{\text{LO}}$ and $\omega_{\text{TO}}$ are the LO and transverse optical (TO) phonon frequencies, respectively. In Eq. (2b), $n$, $m^*$, and $\epsilon_\infty$ are the electron density, effective mass, and dielectric constant, respectively. It is apparent from Eqs. (1), (2a), and (2b) that, in the absence of the photogenerated carrier ($n = 0$), $S_{\text{ph}}^{\text{L}+}$ is unity. This situation corresponds to the phenomenon that the phonon does not couple with electrons: the bare LO phonon. The calculated phonon strengths are depicted by the solid curves as a function of square root of electron density in Fig. 5(a). Note that the electron densities corresponding to the pump-beam powers are estimated as described above. In the present calculation, the following parameters are applied: $\omega_{\text{LO}} = 8.8$ THz; $\omega_{\text{TO}} = 8.0$ THz; $m^* = 0.0665m_0$;[20] $\epsilon_\infty = 10.6\epsilon_0$.[20] At pump-beam power lower than 10 mW, the phonon strength $S_{\text{ph}}^{\text{L}+}$ is almost equal to 1, which means the coupling strength of the LOPC mode is negligibly small. In contrast, at pump-beam power higher than 20 mW, $S_{\text{ph}}^{\text{L}+}$ apparently deviates from 1. At pump beam power of 100 mW, the value of $S_{\text{ph}}^{\text{L}+}$ is decreased to be 0.9, which indicates the considerable deviation of the bare LO phonon. We also note that the value of $S_{\text{ph}}^{\text{L}-}$ amounts to 0.1 at the pump beam power of 100 mW. This calculated result indicates that the plasmonic effect dominates the characteristics of the terahertz wave at pump-beam power higher than about 20 mW; namely, the asymmetric tails of the monocycle signal and coherent LO phonon are attributed to the LOPC modes.

According to the above-mentioned discussion, we analyze the terahertz-wave band taking account of the plasmonic effect above the pump-beam power of 20 mW. In the analysis including the plasmonic effects, we phenomenologically apply the Gaussian functions in order to decompose the Fourier power spectrum. The total number of the Gaussian functions used is four. The four Gaussian functions correspond to the following bands in order of frequency: the surge current band, lower branch LOPC-mode band, the coherent GaAs LO-phonon band, and the upper branch LOPC-mode band. The inset of Fig. 5(b) is the result of the fitting of the terahertz band at the pump-beam power of 100 mW. As shown in the inset, the fitting curves, which are depicted as dashed curves, are in good agreement with the Fourier power spectrum. We plot the peak frequency of the lower branch LOPC-mode band as a closed circle in Fig. 5(b) as a function of the square root of electron density. In Fig. 5(b), the peak frequency of the upper branch LOPC-mode band forming the tail on the higher frequency side of the coherent GaAs LO phonon band is also plotted as an open circle. The calculated frequency-dispersion relations of the lower and upper branches of the LOPC mode are depicted as the solid curves. The positions of the closed and open circles well agree with the dispersion relations of the LOPC mode. The present result indicates that the peak frequency of the observed LOPC-mode band is determined only by the photogenerated carrier density. Thus, we conclude that the LOPC mode is formed in the surge current flowing through the $i$-GaAs

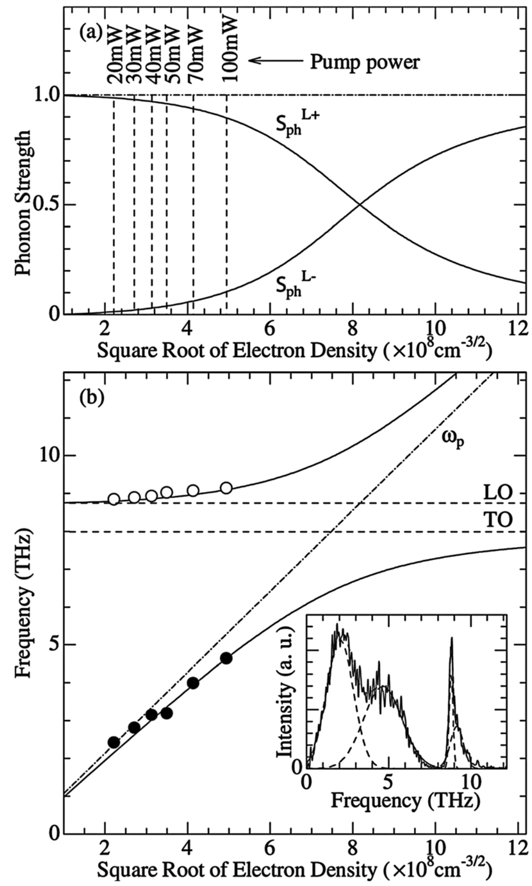


FIG. 5. (a) Phonon strength as a function of square root of electron density calculated using Eq. (1). The two solid curves indicate the phonon strengths of $S_{\text{ph}}^{\text{L}+}$ and $S_{\text{ph}}^{\text{L}-}$. The position of the dashed line corresponds to the square root of the photogenerated electron density at a given pump-beam power. For clarity, the position of $S_{\text{ph}}^{\text{L}+} = 1$ is indicated by the dash-dotted line. (b) The LOPC-mode dispersion relation as a function of the square root of electron density. The solid curve denotes the dispersion curve of the LOPC mode calculated using Eqs. 2(a) and 2(b). The dash-dotted line indicates the plasmon dispersion curve calculated with use of Eq. 2(b). The dashed lines indicate the frequencies of the GaAs TO and LO phonons. The closed and open circles are the peak frequencies of the lower and upper branch LOPC-mode bands estimated by the Gaussian fitting of the Fourier power spectra. In the plot of the circles, the electron densities are estimated from the pump-beam powers. The inset shows the fitting results (dashed curves) of the Fourier power spectrum at the pump-beam power of 100 mW using the four Gaussian functions.

layer; namely, the surge current is not a simple flow of the carriers but involves the collective motion that is the plasmon of the photogenerated carriers. Here, we briefly discuss the reason why the LOPC mode in the underlying *n*-GaAs layer does not contribute to the terahertz wave. Note that the expected frequency of the LOPC mode in the *n*-GaAs layer with a doping concentration of $3 \times 10^{18}$ $cm^{-3}$ is almost equal to the TO-phonon frequency and that such a mode is not observed in the present work. Although the pump beam excites the LOPC mode in the highly-doped *n*-GaAs layer, the background carriers strongly absorb the terahertz wave, which results in a negligibly weak intensity of the terahertz wave relative to that from the *i*-GaAs layer. In fact, according to Ref. 7, the terahertz-wave intensity in an *n*-GaAs crystal with a doping concentration of $2 \times 10^{18}$ $cm^{-3}$ is about 20 times weaker than that in an *i*-GaAs/*n*-GaAs structure. From the viewpoint of the development of the frequency tunable terahertz emitter, the present finding is significant because the frequency of the terahertz wave can be controlled only by the pump-beam power. This is a quite different approach in comparison with Kersting *et al.*[9] and Hasselbeck *et al.*[10] In their studies, the frequency of the terahertz wave from the LOPC mode is determined by the carrier density from dopants.

### B. Disappearance process of the LOPC mode in the *i*-GaAs layer

As described in Sec. II, the relatively high built-in electric field is produced in the *i*-GaAs layer. Consequently, the following hypothesis can be considered: The built-in electric field can instantaneously sweep out the photogenerated electrons from the *i*-GaAs layer to the *n*-GaAs layer and the LOPC mode formed in the *i*-GaAs layer disappears. In order to confirm the above-mentioned hypothesis, we apply the time-partitioning Fourier transform. The time-partitioning Fourier power spectrum $I(\omega)$, where $\omega$ is a frequency, is given by the following equation:

$$I(\omega) = \left| \int_{\tau}^{8ps} A(t) \exp(-i\omega t) dt \right|^2 . \quad (3)$$

Here, $A(t)$ is the terahertz waveform and $\tau$ is the time delay ($-2$ ps $\leq \tau < 8$ ps) determining the time window of the time-partitioning Fourier transform. We note that the time-partitioning Fourier transform has been applied to investigate the time evolution and decay time of the coherent LO phonon and LOPC mode in the optical reflection-type pump-probe measurement.[21–23]

The time-partitioning Fourier power spectra of the terahertz waveform at the pump-beam power of 100 mW are shown in Fig. 6. We performed the fitting with use of two Gaussian functions for the Fourier power spectra at $\tau = -0.3$ ps and $\tau = 0.0$ ps. The two Gaussian functions correspond to the surge current band and the lower branch LOPC-mode band. We note that, as shown in Fig. 3(b), the terahertz waveform shows a rise at $\tau = -0.3$ ps. From the fitting result, the peak frequencies of the lower branch LOPC-mode band, the

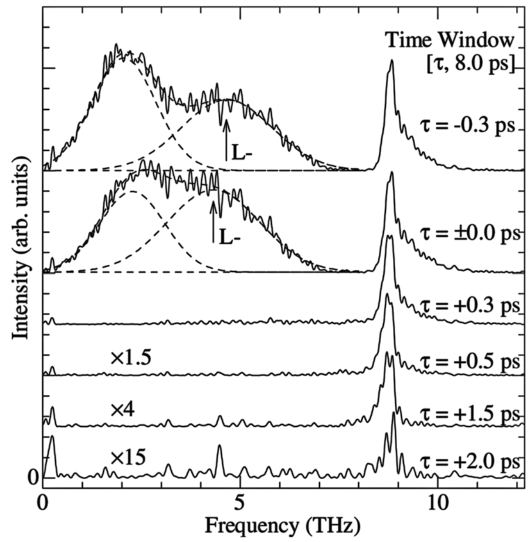


FIG. 6. Time-partitioning Fourier power spectra at various time delays. The dashed line denotes the fitting results using the Gaussian function. The position of the arrow labeled $L_-$ corresponds to the peak frequency of the lower branch LOPC mode obtained by the Gaussian fitting.

position of which is indicated by the arrow, are estimated to be 4.6 and 4.3 THz in order of $\tau$. The present fitting results indicate that the peak frequency of the lower branch LOPC-mode band decreases with an increase of $\tau$. Taking account of the fact that the frequency of the lower branch LOPC mode decreases with a decrease in the electron density, the present fitting result is the apparent evidence for the phenomenon that the photogenerated electrons escape from the *i*-GaAs layer to the *n*-GaAs layer. In addition, at $\tau = +0.3$ ps the bands in the frequency range from 0 to 7 THz, which connect with the monocycle signal, almost disappear. Furthermore, the upper branch LOPC mode, which forms the tail on the higher frequency side of the coherent GaAs LO phonon, almost diminishes. Consequently, it is concluded that the photogenerated electrons escape from the *i*-GaAs layer within 0.6 ps at least. We note that, in the range of $\tau$ from 0.5 to 2.0 ps, only the coherent GaAs LO phonon is observed; namely, the disappearance of the LOPC mode occurs and only the bare coherent GaAs LO phonon emits the terahertz wave. This is the key finding of the present work. Here, we compare the present work with the earlier works. Kersting *et al.*[9] investigated the plasmon mode in *n*-type GaAs crystals. They observed the terahertz wave from the plasmon composed of electrons originating from dopants, so that the concept of the disappearance of the LOPC mode was out of the target in their work. Hasselbeck *et al.*[10] used *n*-type InAs crystals as samples. They reported on the damping effect of the laser-pulse injected carriers on the coherent LOPC mode composed of the background electrons; however, their work was limited within the LOPC mode in the coupling state. Accordingly, the present work is significant in terms of the dynamical coupling characteristics of the LOPC mode formed only by the photogenerated electrons. We note that the photogenerated electrons flow through the intrinsic layer, so that, from the applicational and scientific viewpoints of time-domain terahertz spectroscopy, the present experimental approach is advantageous to investigate the disappearance process of the LOPC mode.

## V. SUMMARY

We have investigated the dynamics of the coupling between the coherent LO phonon and plasmon in the surge current of the instantaneously photogenerated electrons in the *i*-GaAs (200 nm)/*n*-GaAs (3 $\mu$m and $3 \times 10^{18}$ cm$^{-3}$) epitaxial structure using time-domain terahertz spectroscopy. Initially, we verified the formation of the built-in electric field in the *i*-GaAs layer of the *i*-GaAs/*n*-GaAs epitaxial layer with use of both the numerical simulation and the photoreflectance measurement. Next, we performed the terahertz-wave measurements of the *i*-GaAs/*n*-GaAs epitaxial structure from the low density excitation regime to the high excitation regime. The LOPC mode is found from the terahertz-wave measurement. It has been confirmed that the frequency of the LOPC mode is determined only by the pump-beam power. This fact demonstrates that the LOPC mode is formed in the *i*-GaAs layer; namely, the surge current has a plasmon component that couples with the LO phonon. In addition, we performed the time-partitioning Fourier transform in order to investigate the dynamical change of the LOPC mode. Using this analysis, it has been found that the coherent LOPC modes disappear immediately within the time delay of 0.6 ps. After the disappearance of the LOPC mode, only the bare coherent GaAs LO phonon dominates the terahertz waves. The above-mentioned fact indicates that the disappearance process of the LOPC mode is observed in the present study.

## ACKNOWLEDGMENTS

H. T. acknowledges the Grant-in-Aid for Young Scientists B (No. 22760010) from the Japan Society for the Promotion of Science.

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