Terahertz radiation from the coherent longitudinal optical phonon-plasmon coupled mode in an $i$-GaAs/$n$-GaAs epitaxial structure

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Abstract. We have investigated the time-domain terahertz radiation from an undoped GaAs/$n$-type GaAs ($i$-GaAs/$n$-GaAs) epitaxial structure using an optical gating method with a femtosecond pulse laser. We have found from the Fourier power spectra of the terahertz waveforms that two terahertz bands, the frequencies of which obviously depend on the pump power, appear in a high density excitation regime in addition to a broad band due to a surge current and a sharp band due to the coherent longitudinal optical (LO) phonon. Based on a model for the LO phonon-plasmon coupled (LOPC) mode, we reasonably explain the pump-power (photogenerated-carrier-density) dependence of the frequencies of the two bands peculiar to the high density excitation. This fact demonstrates that the two terahertz bands are assigned to the lower and upper branches of the LOPC mode. The frequencies of the LOPC mode are determined only by the pump power. Thus, the LOPC mode originates from the $i$-GaAs layer; namely, the plasmon component of the surge current flowing through the $i$-GaAs layer couples with the coherent LO phonon.

1. Introduction

The generation of terahertz electromagnetic waves has attracted much attention from aspects of physics of ultrafast phenomena and applications in terahertz spectroscopy, sensing, and imaging [1]. There are two well-known generation mechanisms of terahertz waves from a semiconductor surface with use of illumination of a femtosecond pulse laser. One is a surface surge current due to instantaneously generated carriers, which produces a broad terahertz spectrum [2-4]. The other is a coherent optical phonon, which shows a sharp band [5-9]. Here, we emphasize that the surge current 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. Thus, it is expected that the plasmon component of the surge current couples with the coherent longitudinal optical (LO) phonon: the formation of the LO phonon-plasmon coupled (LOPC) mode. However, little has been known about the terahertz radiation from the LOPC mode.

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In the present work, using time-domain terahertz spectroscopy, we have investigated the LOPC mode in an undoped GaAs/n-type GaAs \((i\text{-GaAs}/n\text{-GaAs})\) epitaxial structure. The frequency spectrum was obtained from the Fourier transform (FT) of the time-domain signal. It is found that two terahertz bands appear in a high density excitation regime. The frequencies of the two bands obviously depend on the pump power. The pump-power (photogenerated-carrier-density) dependence of the frequencies of the two bands is reasonably analyzed by a model for the LOPC mode, and the two bands are assigned to the lower and upper branches of the LOPC mode. This fact demonstrates that the LOPC mode is instantaneously formed in the \(i\text{-GaAs}\) layer; namely, the surge current flowing through the \(i\text{-GaAs}\) layer has a plasmon component that couples with the coherent LO phonon. Here, we note that the present result is quite different from the earlier works by Kersting et al. \([10]\) and by Hasselbeck et al \([11]\). The earlier works reported on the LOPC mode in \(n\)-type GaAs and InAs crystals using terahertz spectroscopy. However, it was concluded that the frequency of the LOPC mode is determined only by a background carrier concentration originating from dopants. Thus, the present results of the terahertz radiation from the LOPC mode are peculiar to the \(i\text{-GaAs}/n\text{-GaAs}\) epitaxial structure.

2. Experimental

The sample was an \(i\text{-GaAs}(200 \text{ nm})/n\text{-GaAs}(3 \text{ µm and } 3\times10^{18} \text{ cm}^{-3})\) epitaxial structure grown on a semi-insulating (001) GaAs substrate by metal organic vapor phase epitaxy, where the values in the parentheses denote the individual layer thickness and doping concentration. According to Ref. \([12]\), the surface Fermi level pinning produces a built-in electric field, which was theoretically estimated to be 35 \(\text{kV/cm}\), in the \(i\text{-GaAs}\) layer. Thus, the \(i\text{-GaAs}\) layer was completely depleted, and photogenerated carriers were efficiently swept out from the \(i\text{-GaAs}\) layer by the built-in electric field, which is advantageous to the flow of the surge current.

The signals of the time-domain terahertz waves were measured using laser pulses with a duration time of \(~60 \text{ fs}\). The repetition rate of the laser pulse was \(~90 \text{ MHz}\). The pump beam was focused on the sample with an incidence angle of 45°. The diameter of the spot on the sample surface was about 120 \(\text{µm}\). The photon energies of the pump and gate beams were the same: 1.57 eV. The absorption

![Figure 1](image.png)

Figure 1. (a) Terahertz waveforms at the pump power of 100 mW, which is the highest power in the present work, in the \(i\text{-GaAs}/n\text{-GaAs}\) sample and a bulk (001) GaAs crystal as a reference. (b) Fourier power spectra of the terahertz waveforms.
coefficient and refractive index for GaAs of the present pump beam are $1.4 \times 10^4 \text{ cm}^{-1}$ and 3.69, respectively [13]. 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 $1.2 \times 10^{17} \text{ cm}^{-3}$. The emitted terahertz wave was detected by an optically gated dipole antenna with a gap of 6 μm 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 5% during the measurement under a nitrogen-gas-purge condition.

3. Results and discussion

Figure 1(a) shows the terahertz waveforms at the pump power of 100 mW, which is the highest power in the present work, in the $i$-GaAs/$n$-GaAs sample and a bulk (001) semi-insulating GaAs crystal as a reference. The monocycle signal, which is attributed to the surge current component, is observed around the delay time of 0 ps. Furthermore, the terahertz waveforms exhibit oscillatory patterns. In order to clarify the components of the terahertz waveform, we treat the Fourier power spectrum in frequency domain, which is shown in Fig. 1(b). It is obvious that the profile of the Fourier power spectrum in the $i$-GaAs/$n$-GaAs sample is much different from that in the bulk GaAs crystal. The Fourier power spectrum in the bulk GaAs crystal consists of two bands. One is a broad band peaking at ~2 THz due to the monocycle signal. The other is a sharp band at 8.8 THz due to the coherent LO phonon because 8.8 THz just agrees with the frequency of the GaAs LO phonon. This is an ordinary Fourier power spectrum of the terahertz waveform emitted from a semiconductor surface. In the $i$-GaAs/$n$-GaAs sample, the Fourier power spectrum exhibits a complicated profile. We phenomenologically decompose the Fourier power spectrum with four Gaussian functions. The dashed curves in Fig. 1(b) show the fitted results, which reasonably explain the spectrum profile. The two bands indicated by the solid and open circles newly appear in addition to the two bands that are observed in the bulk GaAs crystal. Thus, the newly appearing two bands are peculiar to the $i$-GaAs/$n$-GaAs sample and to the high density excitation condition.

In order to reveal the origin of the two bands, we focus on the pump-power dependence of the Fourier power spectrum of the terahertz waveform in the $i$-GaAs/$n$-GaAs sample, which is shown in Fig. 2. The broad band shows an asymmetric shape with a tail extending to the high frequency side at the pump powers higher than 20 mW, whereas the peak frequency is almost fixed at about 2 THz. At the pump powers of 70 and 100 mW, the broad band clearly splits into two components. The higher

![Figure 2. Pump-power dependence of the Fourier power spectrum of the terahertz waveform in the $i$-GaAs/$n$-GaAs sample.](image-url)
frequency component corresponds to the terahertz band indicated by the solid circle in Fig. 1(b). In addition, the band of the coherent GaAs LO phonon has an asymmetric tail toward the higher frequency side at the pump powers higher than 20 mW. The asymmetry becomes remarkable with an increase in pump power. The pump-power dependent phenomena described above suggest that photogenerated carriers modify the terahertz waveform; namely, the formation of the LOPC mode is expected.

The frequencies of the lower and upper branches of the LOPC mode, $\omega_{L-}$ and $\omega_{L+}$, are given by [14]

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

where $\omega_{LO}$ and $\omega_{TO}$ are the LO and transverse optical (TO) phonon frequencies, respectively. The plasmon frequency is $\omega_p$ written by

$$\omega_p = \sqrt{\frac{ne^2}{\varepsilon\varepsilon_0 m^*}},$$

where $n$, $m^*$, and $\varepsilon_0$ are the electron density, effective mass, and dielectric constant, respectively. We note that the plasmon of holes does not produce the well-defined lower and upper branches of the LOPC mode described by Eq.(1) because of large damping of the hole plasmon mainly due to the low mobility [15]. Thus, we neglect the contribution of the hole plasmon in this case. In the calculation, the values of the parameters are taken from those of the bulk GaAs crystal [16]. Figure 3 shows the peak frequencies of the two pump-power dependent terahertz bands (solid and open circles), which are observed at pump powers from 20 to 100 mW in Fig. 2, as a function of square root of electron density, where four Gaussian functions were used to resolve the Fourier power spectrum as shown in Fig. 1(b). The electron density was evaluated from the pump power as described above. The solid curves in Fig. 3 depict the calculated dispersion relations of the lower and upper branches of the LOPC mode using Eq.(1). From Fig. 3, it is evident that we detect the terahertz radiation from the LOPC mode. Furthermore, the present result indicates that the peak frequency of the observed LOPC mode is determined only by the photogenerated carrier density. Thus, we conclude that the LOPC mode is
formed in the $i$-GaAs layer; namely, the surge current through the $i$-GaAs layer involves the plasmon component that couples with the coherent LO phonon.

Finally, we discuss the dynamics of the LOPC mode from the viewpoint of the terahertz waveform. In order to eliminate the monocycle signal in the terahertz waveform, we performed the inverse FT of the FT spectrum in the frequency region from 8 to 12 THz at the pump power of 100 mW. The dashed curve in Fig. 4 shows the inverse FT terahertz waveform. We use the following equation, which consists of the oscillation terms of the coherent LO phonon and the upper branch of the LOPC mode, to fit the terahertz waveform:

$$A(t) = A_{LO} \exp\left(-t/\tau_{LO}\right)\cos(\omega_{LO}t + \phi_{LO}) + A_{L+} \exp\left(-t/\tau_{L+}\right)\cos(\omega_{L+}t + \phi_{L+}),$$

where $A$, $\tau$, and $\phi$ are the amplitude, lifetime, and initial phase, respectively. The fitted terahertz waveform using Eq. (3) is depicted by the solid curve in Fig. 4. In the fitting process, we fix the values of the initial phases ($\phi_{LO}$ and $\phi_{L+}$) to zero, assuming the generation mechanism that is instantaneous screening of the surface electric field [17], which is the major generation mechanism of the coherent LO phonon in polar semiconductors such as GaAs. It is evident that the terahertz waveform is well reproduced by the fitting. The ratio of $A_{LO}$ to $A_{L+}$ is estimated to be 1:1.3. The important parameters are the lifetimes of the LOPC mode and coherent LO phonon. The evaluated values of $\tau_{L+}$ and $\tau_{LO}$ are 0.3 and 1.1 ps, respectively. The ultrashort lifetime of the LOPC mode indicates that the photogenerated carriers including the plasmon component in the $i$-GaAs layer are very efficiently swept out by the built-in electric field. In contrast, the coherent LO phonon continues to oscillate after the disappearance of the LOPC mode. This suggests that the LOPC mode and coherent LO phonon are spatially separated.

4. Conclusions

We have detected the time-domain terahertz radiation from the LOPC mode in the $i$-GaAs (200 nm)/$n$-GaAs (3 µm and $3 \times 10^{18}$ cm$^{-3}$) epitaxial structure using the optical gating method. The observed frequencies of the lower and upper branches of the LOPC mode are reasonably explained by the calculated dispersion relations of the LOPC mode. In addition, the frequencies of the LOPC mode depend only on the pump power (photogenerated carrier density). These facts demonstrate that the LOPC mode is instantaneously formed in the $i$-GaAs layer; namely, the surge current flowing through the $i$-GaAs layer has a plasmon component that couples with the coherent LO phonon. Furthermore, the analysis of the terahertz waveform indicates that the lifetime of the LOPC mode is ultrashort: 0.3
ps. Such an ultrashort lifetime results from the fact that the sweep-out of photogenerated carriers in the $i$-GaAs layer is ultrafast due to the built-in electric field peculiar to the $i$-GaAs/$n$-GaAs structure.

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