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Carrier mobility in a polar semiconductor measured by an optical pump-probe technique

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Ultrafast dephasing of the plasmonlike longitudinal optical phonon-plasmon coupled (LOPC) mode in highly doped $n$-GaAs has been investigated by using a femtosecond optical pump-probe technique with 40 THz bandwidth as a function of photodoping levels. The direct measurement of plasmon damping with the help of a wavelet analysis enables us to extract carrier (electron) mobility, which decreases with increasing the photodoping levels. It is found that the mobility is suppressed at high photodoping levels due to electron-hole scattering, while it is enhanced near a critical density, being plausibly attributed to the strong coherent coupling of the LO phonon with the plasmon. © 2009 American Institute of Physics. [DOI: 10.1063/1.3103275]

Very recently, Huber et al. have revealed ultrafast build-up of the LOPC modes in InP by using optical pump-terahertz probe spectroscopy, however, relaxation dynamics of the LOPC modes and carrier mobility have not been examined.

In this letter, the direct measurement of carrier mobility in polar-semiconductor is proposed using a femtosecond optical pump-probe technique. Relaxation dynamics of strongly damped LOPC modes are observed in time-frequency space, which enables us to extract $\tau$. By using $\tau$, the electron mobility in $n$-type semiconductor is obtained at different photodoping levels, and found that the mobility is suppressed at high photodoping levels due to electron-hole scattering, while it is significantly enhanced near the critical carrier density.

To detect the plasmonlike LOPC modes in real time domain, we use an electro-optic detection combined with a standard pump-probe method utilizing a mode-locked Ti:sapphire laser with a pulse duration of 15 fs, and a center wavelength of 815 nm ($=1.52$ eV). This ultrashort pulse laser enables us to detect optical responses over 40 THz bandwidth. The average power of pump-beam was varied from 10 to 200 mW, while that of the probe was kept at 4 mW. The maximum photoexcited carrier density by the pump light was estimated to be $N_{\text{exc}}=1.8\times10^{18}$ cm$^{-3}$ from the pump power density and the absorption coefficient. The sample used was Si-doped $n$-type GaAs with a doped carrier density of $N_{\text{dep}}=1.0\times10^{18}$ cm$^{-3}$. In our experimental condition, the total density of electron plasma is larger than that of hole plasma, and thus the phonon-plasmon coupling is dominated by photogenerated electron plasma rather than photogenerated hole plasma. In addition to this effect, the relaxation of the photogenerated holes is extremely fast ($\approx 75$ fs), which would make hole plasma difficult to couple with the LO phonons. The transient anisotropic reflectivity change ($\Delta R_{eo}/R$) was recorded as a function of the time delay at room temperature.

The time derivatives of $\Delta R_{eo}/R$ signal for $n$-GaAs at the two different photoexcited carrier densities are shown in Fig. 1. The coherent oscillations with a strong mode beating are clearly observed, which is due to the coexistence of the LO phonon (8.75 THz), and the lower ($L_-$) and the upper ($L_+$) branches of the LOPC modes. As shown in the Fourier

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transformed (FT) spectra in the inset of Fig. 1(a), the peak frequency of the $L_-$ and the $L_+$ modes at $N_{\text{exc}}=1.8\times10^{18}\text{ cm}^{-3}$ are 7.9 and 19 THz, respectively. On the other hand, at the lowest limit of $N_{\text{exc}}=8.9\times10^{16}\text{ cm}^{-3}$ they are 7.9 and 15 THz, respectively, as shown in Fig. 1(b).\textsuperscript{10,11} Obviously, the fit of the time-domain data using exponentially damped harmonic oscillations are difficult because of the complicated coherent oscillations appearing at the early time delay of <300 fs.

In order to explore the time-frequency dynamics of coupled plasmon-phonon system, a wavelet analysis\textsuperscript{12} was applied to the time-domain data in Fig. 1. Figure 2 shows three-dimensional image of electro-optic response obtained by the wavelet analysis. At the high photodoping level [Fig. 2(a)], the transient frequency of the $L_+$ mode (23 THz) is higher than that observed in the static FT spectra (19 THz), and it decays more quickly than those of the LO and the $L_-$ modes, which generate mode-beating patterns at 8–9 THz, living until a few picoseconds after photoexcitation. The higher frequency of the $L_+$ mode (23 THz) than that observed in the static FT in Fig. 1 (19 THz) is possibly due to transient nonequilibrium electron distribution (hot electrons),\textsuperscript{1} which has much higher electron velocity than that of the cold plasma accommodating at the bottom of the conduction bands.\textsuperscript{12} At the low photodoping level [Fig. 2(b)], on the other hand, the transient frequency of the $L_+$ mode is only slightly higher (16 THz) than that observed in the static FT spectra (15 THz). In this lowest photodoping limit, the LOPC dynamics would be governed by the cold plasma, and thus the transient frequency of the $L_+$ mode does not shift much compared to that observed in the static FT spectra. It is to be noted that the frequency of the $L_+$ mode (15 THz) matches to that observed by Raman measurement at the similar carrier doping level.\textsuperscript{15}

In order to extract the relaxation time of the plasmonlike LOPC mode, the peak intensity of the mode is plotted as the function of the time delay at three different photodoping levels at the fixed majority carrier density of $N_{\text{dop}}=1.0\times10^{16}\text{ cm}^{-3}$, as shown in Fig. 3(a). It is noticed that the rise time of the peak significantly depends on the photodoping levels. It decreases from 120±15 to 73±15 fs as the photodoping increases. This behavior is similar to the carrier density dependence of the buildup time of LOPC modes observed in photoexcited InP, and would be explained by the fastest corrective response time of the many-body system.\textsuperscript{6}

The current interest is the relaxation time rather than the build-up time. The relaxation time $\tau$ of the plasmonlike LOPC mode also depends on the photodoping levels. The value of $\tau$ decreases from 135±15 to 79±15 fs as the photodoping increases, which is the similar effect observed in InN\textsuperscript{14} and in the slightly low photodoped n-GaAs.\textsuperscript{10} Since the relaxation time of the plasmonlike LOPC mode can be directly corresponded to the carrier relaxation time $\tau$ under the condition that the dephasing of the plasmonlike LOPC mode ($L_+$ mode) is governed by the damping of the plasmon rather than that of the LO phonon,\textsuperscript{10,15} one can calculate the carrier mobility by $\mu=e\tau/m^*$. Here, we assume that there is no scattering of electrons into X or L valleys and the parabola conduction band structure at $\Gamma$ point, in which the standard value of the electron effective mass ($m^*=0.067m_e$) can be used.\textsuperscript{1}

Figure 3(b) shows the carrier (electron) mobility extracted by using $\mu=e\tau/m^*$, where $e=1.602\times10^{-19}\text{ C}$ and $m_e=9.1\times10^{-31}\text{ kg}$. The mobility exhibits monotonic decrease with increasing the carrier density with a saturation at

![Image](image_url)
the higher density than \( N_{\text{tot}} \approx 1.8 \times 10^{18} \text{ cm}^{-3} \), where \( N_{\text{tot}} = N_{\text{dep}} + N_{\text{exc}} \). Since we are changing the photodoping levels only, enhanced impurity scattering or temperature-dependent polar phonon (Fröhlich) scattering cannot account for the change in the mobility. Hence, the increased density of carriers plays a dominant role in that change. The mobility can be fit well to the empirical Caughey–Thomas relation in which all the scattering mechanisms are empirically included.\(^1\)

\[
\mu = \frac{\mu_{\text{max}} - \mu_{\text{min}}}{1 + (N_{\text{tot}}/N_{\text{ref}})^{\alpha}} + \mu_{\text{min}},
\]

where \( N_{\text{ref}} \) and \( \alpha \) denote fitting parameters, and \( N_{\text{ref}} = 1.26 \times 10^{18} \text{ cm}^{-3} \) and \( \alpha = 13.2 \) are obtained. Although the model describe the behavior only qualitatively, the relatively strong decrease in the mobility at lower density than \( N_{\text{tot}} \approx 1.8 \times 10^{18} \text{ cm}^{-3} \) would be dominated by electron-hole scattering.\(^9\) The saturation of the mobility at higher density would come from the slow down of the carrier relaxation due to reabsorption of carriers by the high density hot phonons\(^2\) or screening effect of Coulomb interactions.\(^3\)

When we compare the mobility obtained in the present study with that by Hall measurement (2300 cm\(^2\)/Vs), it is found that the drift mobility \( \mu \) determined by the coherent phonon spectroscopy is higher than the Hall mobility at \( N_{\text{tot}} \leq 1.4 \times 10^{18} \text{ cm}^{-3} \). Since the observed LOPC modes are always plasmonlike at \( N_{\text{tot}} \approx 1.0 \times 10^{18} \text{ cm}^{-3} \),\(^4\) transformation of the LOPC modes from plasmonlike into phononlike would play negligible role in the enhanced mobility at \( N_{\text{tot}} \leq 1.4 \times 10^{18} \text{ cm}^{-3} \). There are a few plausible explanations for the enhanced electron mobility: (i) the coherent coupling of LO phonon and the plasmon through Coulomb interactions\(^4\) and (ii) the difference between the two experimental methods. Regarding to the reason (ii) above, the Hall mobility \( \mu_{\text{H}} \) can generally be expressed by \( \mu_{\text{H}} = r_{\text{H}} \mu_{\text{i}} \), where \( r_{\text{H}} \) is the Hall ratio (or Hall factor).\(^1\) The value of \( r_{\text{H}} \) depends on the scattering mechanisms that contribute to the relaxation time \( \tau \). In our case (Fig. 3), \( r_{\text{H}} \) varies from \( r_{\text{H}} \approx 1.0 \) at \( N_{\text{tot}} \approx 1.4 \times 10^{18} \text{ cm}^{-3} \) to \( r_{\text{H}} \approx 1.15 \) at \( N_{\text{tot}} \approx 1.8 \times 10^{18} \text{ cm}^{-3} \). These facts signify that the scattering mechanisms contributing to \( \tau \) in the present study is significantly different from that of the simple electron gas drifting under the weak and static electric field found in Hall measurements. The main scattering mechanism of nonequilibrium electron gas is governed by electron-hole scattering,\(^2\)\(^6\) while that of the simple (equilibrium) electron gas is affected by impurity and polar phonon (Fröhlich) scatterings as well as carrier-carrier scattering. Regarding to the reason (i) above, the enhanced carrier mobility observed by the coherent spectroscopy implies that one may manipulate the carrier mobility by controlling the Coulomb interaction via coherent control of the LOPC modes.\(^2\)

It should be noted that the proposed method is valid only if the coupled mode is completely plasmonlike whose dephasing is determined by the plasmon damping.

In conclusion, the carrier (electron) mobility was determined from the dephasing time of the plasmonlike coherent LOPC mode \( \langle L_+ \rangle \) in \( n \)-GaAs, which is obtained by mapping the time-frequency dynamics of the LOPC modes by the use of the wavelet analysis. The electron mobility extracted from the coherent phonon spectroscopy decreases with increasing the photodoping levels, indicating the suppression of the mobility by enhanced electron-hole scattering. The availability of this technique will spread over the polar semiconductors, such as SiC and GaN, under the condition that the photodoping level assures that the LOPC mode is plasmonlike.

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11. The main difference between the previous experiment (Ref. 10) and the present experiment is the mechanical detection (<30 THz in the previous one), which enabled us to observe plasmonlike LOPC modes at the higher total carrier density than those in the previous study.
17. The data in Fig. 3(b) was fit to also the power law of \( \mu \sim N_{\text{dep}}^{-3/2} \) and \( \mu \sim N_{\text{tot}}^{1/2} \), according to the Drude model and the Coulomb screening model, respectively (Ref. 14). The former model reproduced the reduction in \( \mu \) at \( N_{\text{dep}} \lesssim 1.4 \times 10^{18} \text{ cm}^{-3} \) well, while the latter one explained the behavior of \( \mu \) at \( N_{\text{tot}} \gtrsim 1.4 \times 10^{18} \text{ cm}^{-3} \).