Ultrafast decay of coherent plasmon-phonon coupled modes in highly doped GaAs

Muneaki Hase,* Shin-ichi Nakashima, Kohji Mizoguchi, and Hiroshi Harima
Department of Applied Physics, Osaka University, Yamadaoka 2-1, Suita 565-0871, Japan

Kiyomi Sakai
Kansai Advanced Research Center, CRL, 588-2 Iwaoka, Nishi-ku, Kobe 651-2401, Japan
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We report on the ultrafast decay of coherently excited plasmon-phonon coupled modes in a highly doped *n*-GaAs. Coherent oscillations of the upper branch (L_+) as well as the lower branch (L_-) of the coupled modes have been observed by a femtosecond pump-probe technique with 20 fs ultrashort laser pulses. The decay time of the L_+ mode estimated from a time partitioning Fourier transform spectra reveals that the decay rate increases linearly with increasing photoexcited carrier density. This result leads to a conclusion that the ultrafast decay of the coherent L_+ mode is mainly caused by loss of coherence in electron-hole plasmas when the photoexcited carrier density is higher than doping levels. [S0163-1829(99)01048-6]

I. INTRODUCTION

Recently, the progress of ultrashort pulse lasers has led to new techniques such as time-domain spectroscopy of the terahertz (THz) oscillations in solids. There have been a large number of reports on the observation of coherent lattice vibrations in semiconductors,[1–6] in semimetals,[7–10] and coherent molecular vibrations in molecular crystals.[11] These techniques have made it possible to obtain not only the amplitude but also the phase of coherent phonon oscillations. Coherent LO phonons in GaAs were observed first by Cho *et al.*[1] and are thought to be generated by a sudden screening of the surface space-charge field due to the increase in photoexcited carrier density.[2] This generation mechanism was experimentally supported by Dekorsy *et al.*, who reported on the direct observation of the ultrafast transient screening and the associated charge-carrier transport in a (100)-oriented GaAs surface.[3] Their results showed that the surface space-charge field in the depletion region of samples was strongly coupled with the polarization field of the LO phonons.

The LO phonon-plasmon coupled (LOPC) mode has mainly been studied by Raman scattering spectroscopy, i.e., frequency-domain spectroscopy.[12,13] The relaxation dynamics of the coherent phononlike LOPC mode in *n*-doped GaAs (in the range $10^{16}-10^{17}$ cm^{-3}) have been examined using time-resolved coherent anti-Stokes Raman scattering (CARS), and a large increase of the measured dephasing rate with electron density was demonstrated for electron densities larger than 10^{16} cm^{-3}. This result provides evidence of carrier-induced dephasing of the phononlike mode.[14] The coupled mode is also expected to be observed by the time-domain spectroscopy.

Kuznetsov and Stanton have developed a microscopic theory of the plasmon-phonon oscillations that are generated by ultrafast optical excitation in GaAs, and they have discussed the possibility of observing LOPC modes in the time domain.[15] These modes have been investigated by a pump-probe technique, i.e., time-domain spectroscopy. Recently, the coherent LOPC mode (L_- mode) has been observed in *n*-GaAs by Cho *et al.* using 50 fs laser pulses.[4,5] They reported that the background majority plasma was involved in the coherent LOPC mode, and that the frequency of the coupled modes was determined by the total electron density that resulted from both background doping and optical excitation. Subsequently, we have succeeded in observing the coherent L_+ mode in GaAs and demonstrated that the line shape and frequency of the L_- and L_+ bands vary strongly with photoexcited carrier density.[16] Little is known, however, about the dynamics of the plasmonlike coherent L_+ mode. A systematic study on the ultrafast decay of the coupled modes in a wide range of the carrier density is required for understanding the nature of coherent LOPC modes.

In this paper, direct observation of the ultrafast dynamics for the coherent plasmon-phonon coupled modes in highly doped *n*-GaAs is conducted using a time-resolved pump-probe technique and a 20 fs ultrashort pulse laser. The decay time of the plasmonlike L_+ mode in a high carrier density region is determined by means of a time partitioning Fourier transform (TPFT). The measurement of the decay times for different electron densities suggests that the ultrafast decay of the L_+ mode for the electron densities smaller than 1×10^{18} cm^{-3} is mainly caused by loss of coherence due to electron-hole scattering in the presence of electron-hole plasma generated by optical excitation.

II. ULTRAFAST RELAXATION OF COHERENT LO-PHONON PLASMON COUPLED MODES

Here we will briefly describe a theoretical model for relaxation of the LOPC modes in the femtosecond time domain. Using equations of motion for phonons and electrons, Vallée *et al.* derived the decay rate of the L_+ modes $1/T_2^+$ as follows:[14]

$$\frac{1}{T_2^+}=\frac{1}{T_2^0}+\frac{\omega_p^2(\omega_{LO}^2-\omega_0^2)}{2\omega_{LO}^4\langle\tau\rangle_\infty}=\frac{1}{T_2^0}+\frac{2\pi e^2 n_e(\omega_{LO}^2-\omega_0^2)}{\epsilon_\infty m^*\omega_{LO}^4\langle\tau\rangle_\infty}, \tag{1}$$

where $1/T_2^0$ is the decay rate of the bare LO phonon, ω_p the plasma frequency, ω_{LO} the LO phonon frequency, ω_0 the TO phonon frequency, e the charge of electron, n_e the electron density, ϵ_∞ the dielectric constant, m^* the reduced mass of the electron, and $\langle\tau\rangle_\infty$ the average electron momentum scattering time for the high-frequency (dynamic) limit ($\omega\tau \gg 1$, where ω is the collective electron oscillation frequency and τ the velocity-dependent electron momentum relaxation time used in the Drude model).[14] The electron momentum relaxation time τ is originated from the effect of all electron scattering processes, i.e., electron-hole scattering, electron-acoustic phonon scattering and by spatial disorder (impurities and defects). Equation (1) shows that the net decay rate is related to a phonon damping (first term) and a plasmon damping (second term). It is expected that the decay rate of the coherent L_+ mode varies with the electron density n_e and the average electron momentum scattering time $\langle\tau\rangle_\infty$ for highly doped *n*-GaAs. The average electron momentum scattering time $\langle\tau\rangle_\infty$ was estimated to be 55 ~85 fs for the electron density of 10^{16}–10^{17} cm^{-3},[14] and it is expected to be also below 100 fs for electron density larger than 10^{17} cm^{-3} under our experimental conditions. In addition to the electron momentum scattering, dynamical carrier-carrier scattering events occur in the conduction bands because nonequilibrium carriers are generated by optical excitation. Equation (1) has been established for low carrier density, below 10^{17} cm^{-3}. However, we have treated Eq. (1) as an approximate expression to examine the dynamics of the L_+ mode for the electron density around 10^{18} cm^{-3}.

III. EXPERIMENTS

The measurements were carried out by a reflection-type pump-probe technique at room temperature. The light source was a mode locked Ti:sapphire laser operating at a wavelength of 800 nm with a pulse duration of 20 fs. The energy of the laser pulse was 1.55 eV, being above the band gap of GaAs at 300 K (=1.43 eV), and electron-hole pairs were photoexcited. The pump-beam power was varied by a neutral density filter from 20 to 100 mW, while the probe-beam power was fixed at about 2 mW. The pump and probe beams were focused on *n*-type GaAs (100) samples to a diameter of about 100 μm for the pump beam and about 50 μm for the probe beam. To minimize spatial inhomogeneity of the photoexcited carrier density, only the center of the pump-beam spot was monitored by the probe beam with smaller spot size. The pump beam was mechanically chopped at 2 kHz for signal detection by a lock-in amplifier. We used an electro-optic (E-O) sampling technique to measure the surface electric field, which was modulated by the microscopic longitudinal oscillations, i.e., the LOPC mode.[2] The reflectivity change $\Delta R_{eo}/R$ was recorded as a function of the time delay between the pump and the probe pulses.

IV. RESULTS AND DISCUSSION

Figure 1 shows the oscillatory component of $\Delta R_{eo}/R$ for a sample with a doped carrier concentration of $n_{dop}=1\times10^{18}$ cm^{-3} at the photoexcited carrier densities n_{exc} between 2×10^{17} cm^{-3} and 1×10^{18} cm^{-3}. The observed autocorrelation (AC) of the pump and probe pulses shown in this

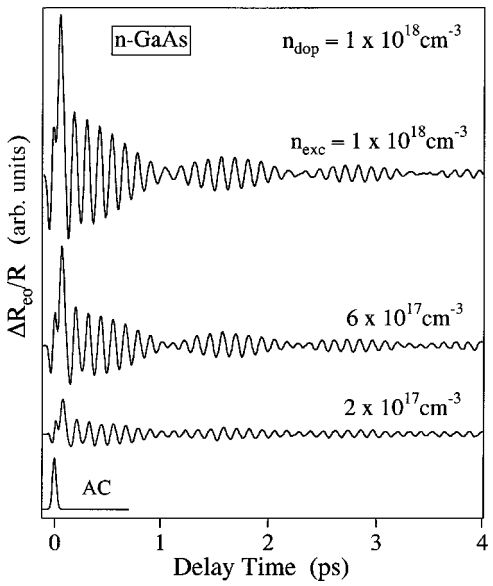

FIG. 1. The time differential of the electro-optic signal for *n*-GaAs ($n_{dop}=1\times10^{18}$ cm^{-3}) at various excited carrier densities. The oscillatory component indicates coherent collective motions of atoms and carriers.

figure indicates that the time resolution (FWHM of AC) is about 35 fs. The photoexcited carrier density n_{exc} was estimated from the pump-beam power density and the absorption coefficient. The total carrier density is given by a sum of the majority electron density and the excited electron density ($n_{dop}+n_{exc}$).[4,16] As shown in Fig. 1, beating signals appear clearly at all values of n_{exc}. The Fourier transform (FT) spectra of these time-domain signals are shown in Fig. 2,

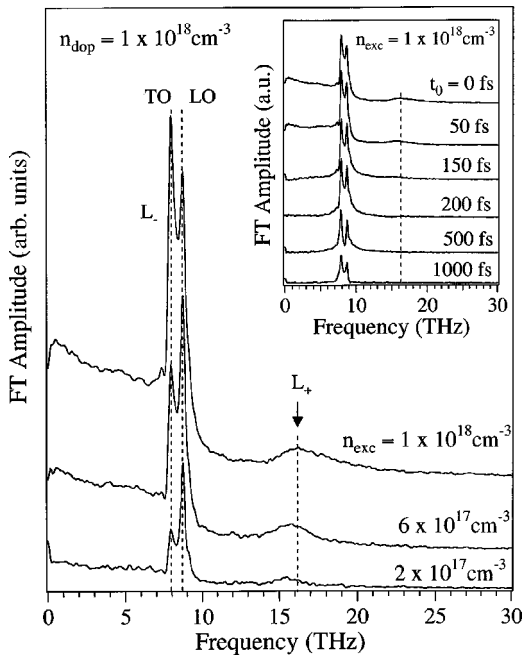

FIG. 2. The Fourier transform spectra obtained from time-domain signals. The frequencies of the bare LO and TO bands are indicated by dotted lines. Both the upper and lower branches of LOPC modes (L_- and L_+, see arrows) were observed. Inset: The time partitioning Fourier transform spectra for $n_{exc}=1\times10^{18}$ cm^{-3} at various t_0's.

where the bare LO- and TO-phonon frequencies (8.76 and 8.06 THz, respectively) are shown by dotted lines. The spectral profiles depend on the excited carrier densities. The dependence is weak for samples with a doping level of $n_{dop}=1\times10^{18}$ cm^{-3} compared to those with a level of $n_{dop}=3\times10^{17}$ cm^{-3}.[16] The upper branch (L_+) of the LOPC band appears distinctly at the high frequency side of the bare LO band, and the lower branch (L_-) appears at a frequency almost the same as that of the TO band. The frequency of the L_- mode does not significantly vary with the carrier density in contrast with that of the L_- mode observed in low doped samples,[16] but the amplitude ratio of the L_- mode to the LO phonon increases as the photoexcited carrier density increases. This fact can be explained by considering that the L_- mode is almost TO phononlike in the highly doped GaAs. The band width of the L_- mode is comparable to that of the LO phonon, showing slow decay of the L_- mode. The frequency of the L_+ mode varied slightly with the photoexcited carrier density. This is because the excitation carrier density is smaller than the majority carrier density and the L_+ mode frequency would be mainly determined by the majority carrier density. The highest frequency of the L_+ mode observed was about 16.0 THz when the total carrier density was 2×10^{18} cm^{-3}. This frequency value is in good agreement with that observed by Raman scattering in a sample with the same doping level,[12] and is also consistent with the prediction based on the microscopic theory of the plasmon-phonon oscillations created by ultrafast optical excitation.[15] It is to be noted that the LO phonon mode is observed at the depletion region and that the inhomogeneous carrier density due to a pump beam profile would affect the line shape of the coherent LO phonon and LOPC modes.[15] This density inhomogeneity would explain the presence of the wing on the high energy side of the LO phonon and the asymmetric shape of the L_+ line.

We estimated the decay time using a time partitioning Fourier transform (TPFT), because the line shape of the LO, L_-, and L_+ modes are asymmetric and it is difficult to deduce the decay time of the coherent LOPC modes by fitting exponentially damped harmonic oscillations to the observed time domain signals.[16] The FT spectra at the delay time t_0 was obtained by the following Fourier transform equation for windows from t_0 to ∞, $[t_0,\infty]$;

$$I(\omega)=\int_{t_0}^{\infty}\left(\frac{\partial\Delta R_{eo}}{\partial t}\right)e^{-i\omega t}dt. \tag{2}$$

Since the coherent oscillations examined here, especially the coherent L_+ mode, are rapidly decaying modes, the TPFT spectra for a time interval $[t_0,\infty]$ can roughly be regarded as the spectrum at the delay time t_0, although the signal includes the oscillatory component in a time region far from t_0. The inset in Fig. 2 shows the TPFT spectra obtained for a sample with a carrier concentration of $n_{dop}=1\times10^{18}$ cm^{-3} and a photoexcited carrier density of $n_{exc}=1\times10^{18}$ cm^{-3}. Comparing the TPFT spectra at different t_0's, we see that the L_+ mode decays rapidly, while the LO and L_- oscillations decay slowly (the time constants are longer than 500 fs). The peak frequencies of the L_+ and L_- modes do not change as t_0 varies, showing that the frequency of the LOPC mode is determined by the initial total carrier density and that the effective carrier density is constant within a few picoseconds. This result reflects that the population-decay (recombination) time of the photoexcited carriers is $\sim$100 ps in GaAs. In Fig. 3, the peak FT amplitudes of the LO phonon, L_+, and L_- modes shown in the inset of Fig. 2 are plotted as a function of t_0. The FT amplitudes of both the LO phonon and L_- mode increase at the initial stage (the rising time is about 50 fs), and then decay exponentially. This rise of the FT amplitude might be due to the finite width of the pump and probe pulses. The decay time obtained by fitting the data to a single exponential decay is 630 ± 40 fs for the LO phonon, 920 ± 40 fs for the L_- mode, and 130 ± 40 fs for the L_+ mode. The decay time of the LO phonon is close to that of semi-insulating GaAs with a photoexcited carrier density of 10^{18} cm^{-3} (700 fs),[1] but is shorter than that of intrinsic GaAs which was obtained by using CARS (about 4.2 ± 0.4 ps at 300 K).[17] This shortening of the LO phonon decay is discussed later.

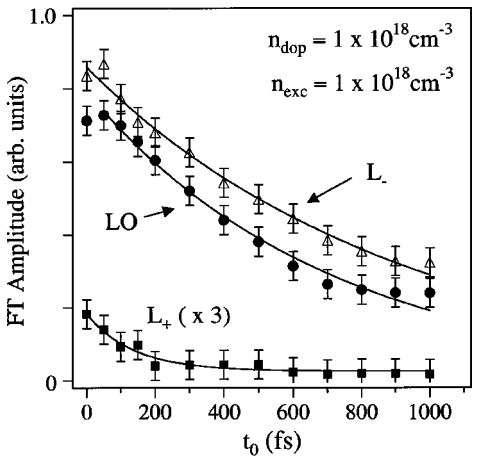

FIG. 3. FT amplitudes as a function of t_0 obtained by the time partitioning Fourier transform. Solid lines are fitted curves with a single exponential decay.

The TPFT spectra for a sample with carrier concentration of $n_{dop}=3\times10^{17}$ cm^{-3} were also obtained by Hase *et al.*[16] The decay times of the L_+ mode obtained for the highly doped sample ($n_{dop}=1\times10^{18}$ cm^3) and the low doped sample ($n_{dop}=3\times10^{17}$ cm^{-3}) are plotted in Fig. 4 as a function of the photoexcited carrier density n_{exc}. The decay time for the highly doped sample is almost independent of n_{exc}, being about 130 ± 40 fs. For the low doped sample, however, the decay time decreased as n_{exc} increased, and reached the same value as that of highly doped sample with $n_{exc}=1\times10^{18}$ cm^{-3}. These results indicate that the decay time of the coherent L_+ mode depends strongly on the photoexcited carrier density n_{exc} when the doping level is less than n_{exc}, while the decay time is almost independent of n_{exc} when the doping level is larger than n_{exc}. The decay time of the L_+ mode (130 ± 40 fs) for the photoexcited carrier density of 1×10^{18} cm^{-3} is much shorter than the LO phonon decay time (about 4.2 ps at 300 K). This would indicate that the coherent L_+ mode observed here is a plasmonlike mode and the second term in Eq. (1) plays an important role in the ultrafast decay of the coherent L_+ mode. Thus, the decrease of the decay time with n_{exc} for the low doped sample can be explained by the increase in damping of the electron-hole

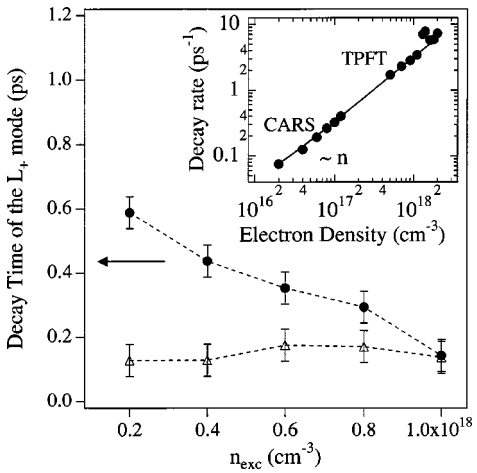

FIG. 4. The decay time of the L_+ mode for highly doped (open square; $n_{dop}=1\times10^{18}$ cm^{-3}) and low doped (closed circle; $n_{dop}=3\times10^{17}$ cm^{-3}) *n*-GaAs. The dotted lines are guides for the eye. The inset shows the decay rates of the L_+ mode as a function of the total electron density obtained by CARS (using the Vallée data) and our TPFT. Our results can be fitted to the line that depends linearly on the electron density.

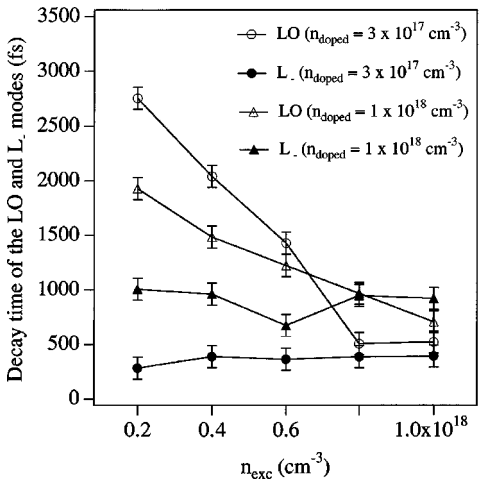

FIG. 5. The decay time of the LO phonon and L_- modes for the low doped and highly doped *n*-GaAs obtained by TPFT methods. These are of the LO phonon (open square), L_- mode (closed square) for the $n_{dop}=1\times10^{18}$ cm^{-3} sample and of the LO phonon (open circle), L_- mode (closed circle) for the $n_{dop}=3\times10^{17}$ cm^{-3} sample.

plasma generated by optical excitation.[18] The constant decay time for the highly doped sample can be explained by the fact that the majority carrier density is larger than the photoexcited carrier density, and the decay rate would be determined by electron scattering with impurities and defects. In the inset of Fig. 4, the decay rates of the L_+ mode are plotted as a function of the total electron (carrier) density. According to the results of CARS experiments by Vallée *et al.*, the decay rate of the phononlike L_+ mode varies linearly with electron density n_e as shown in the inset, in which we plot their data for $2/T_2^+-2/T_2^0$, where $1/T_2^+$ is the decay rate of L_+ mode and $1/T_2^0$ is the decay rate of the bare LO phonon.[14] The decay rates we obtained for the plasmonlike L_+ mode also reveal linear dependence for electron densities smaller than 1×10^{18} cm^{-3}, as shown in the inset, although the decay rate did not show a systematic change for the electron density larger than 1×10^{18} cm^{-3}. The corresponding average electron scattering time $\langle\tau\rangle_\infty$ deduced using Eq. (1) is almost constant ($\sim55\pm10$ fs) for the electron densities from 5×10^{17} cm^{-3} to 2×10^{18} cm^{-3}. The deviation from linear dependence of our TPFT data for the electron density larger than 1×10^{18} cm^{-3} in the inset may be originated from electron scattering by impurities and defects. It has been reported that electron-hole scattering is a dominant scattering process in the presence of photogenerated electron-hole plasma.[18] Their report is consistent with our result that the decay rate of the coherent L_+ mode linearly increases with electron density n_e, as expected from the second term in Eq. (1). Our results together with their report demonstrate the ultrafast decay of the coherent L_+ mode for electron densities below 1×10^{18} cm^{-3} is caused by loss of coherence due to electron-hole scattering in the presence of electron-hole plasma generated by optical excitation.

Portella *et al.* investigated the *k*-space carrier-carrier scattering in GaAs by measuring the transient relaxation of an anisotropic population, using 9 fs ultrashort pulses.[19] In their experiment, the dependence of the momentum relaxation time on carrier density showed that the decay rate varied with $\sim n_e^{1/3}$, and they concluded that carrier momentum is rapidly redistributed by a screened Coulomb interaction between carriers.[19] Our results for the decay rate of the L_+ mode ($1/T_2^+\sim n_e$) deviate from their results ($\sim n_e^{1/3}$), and this fact indicates that the *k*-space carrier-carrier scattering would not affect the ultrafast decay of the L_+ mode, although the *k*-space carrier scattering may partly contribute to dephasing of the L_+ mode. The LO phonon emission would not contribute to the decay of the coherent L_+ modes in our samples, although the decay time of the plasmonlike L_+ mode is close to the LO phonon emission time of photoexcited carriers (about 100 fs).[20,21] Further studies on the electron densities larger than $\sim2\times10^{18}$ cm^{-3} is required to understand the nature of the overdamped coherent L_+ mode.

The dephasing of the coherent LO phonon and the L_- mode has been also studied by using TPFT. As shown in Fig. 5, for both highly doped ($n_{dop}=1\times10^{18}$ cm^{-3}) and low doped ($n_{dop}=3\times10^{17}$ cm^{-3}) samples, the decay times of the coherent LO phonon and the coherent L_- modes are obtained. The LO phonon decay time decreases with increasing the doping density and it decreases as the photoexcited carrier density n_{exc} is increased. This result indicates that the dephasing of the coherent LO phonon observed in our experiment is due to electron-phonon interaction rather than phonon-phonon interaction introduced by crystal potential anharmonicity.[14] The decay time of the coherent L_- mode does not drastically change compared to the LO phonon. The decay time of the L_- mode slightly decreases when n_{exc} is increased in the highly doped sample, whereas, in the low doped sample, the decay time of the L_- mode slightly increases when n_{exc} is increased as shown in Fig. 5. This result suggests that the L_- mode is a phononlike mode in the highly doped sample, and that the L_- mode slightly changes from a plasmonlike to a phononlike mode in the low doped sample as n_{exc} increases.

V. CONCLUSION

Ultrafast decay of the coherent LO phonon-plasmon coupled modes was investigated using a femtosecond pump-probe technique. We observed both the upper and lower branches of the coherent LO phonon-plasmon coupled modes in *n*-GaAs. The coherent L_- mode observed in highly doped GaAs was almost a TO phononlike mode and the frequency did not drastically change. The decay time of the L_+ mode was estimated by a time partitioning Fourier transform spectra for various carrier densities. The decay rate of the L_+ mode was found to vary linearly with electron densities below 1×10^{18} cm^{-3}. This finding suggests that the ultrafast decay of the plasmonlike L_+ mode is caused by loss of coherence due to the damping of electron-hole plasma when the doping level is less than photogenerated carrier density. The ultrafast decay of the coherent L_+ mode would be mainly caused by electron scattering with impurities and defects when the doping level is larger than photogenerated carrier density.

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*Present address: 2nd Research Group, National Research Institute for Metals (NRIM), 1-2-1 Sengen, Tsukuba, 305-0047 Ibaraki, Japan.

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