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Crossover from coherent to incoherent excitation of two-dimensional plasmons in $GaAs/Al_xGa_{1-x}As$ single quantum wells by femtosecond laser pulses

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We have investigated the dynamics of two-dimensional (2D) free carrier plasma oscillations in selectively doped $GaAs/Al_xGa_{1-x}As$ single quantum wells by time-domain terahertz (THz) emission spectroscopy. Considering both excitation energy and power dependence of the 2D plasmon radiation intensity, we have found two distinct excitation mechanisms of the 2D plasmon; coherent impulsive stimulated Raman process and incoherent sudden heating of the electron system by photoexcited carriers. By monitoring the phase of the emitted THz radiation, a clear crossover from one mechanism to the other is observed as the pump photon energy goes through the absorption edge of the electron system.

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Plasma oscillation is a normal oscillation mode of electron systems and is one of the most fundamental elementary excitations in solids. Spectroscopic measurements, such as far infrared (FIR) absorption and Raman scattering, have been extensively carried out and the knowledge of plasmon dispersions has been well established. Since typical plasma frequencies in semiconductors are in the terahertz (THz) range, plasma oscillation has been recognized as a promising candidate for solid-state THz sources. In particular, plasma oscillations of two-dimensional (2D) electrons accumulated in semiconductor heterostructures as well as in Si metal-oxide-semiconductor structures[1,2] have been extensively studied and, indeed, quasi-cw FIR emission from plasmons thermally excited by current has been observed.[3–6] However, all these works are focused on the investigation of quasi-equilibrium properties of plasmons. The understanding of dynamical properties of electron plasma oscillations has yet to be established.

Recent progress in femtosecond laser technology has made it possible to study the dynamics of elementary excitations such as electronic motions and lattice vibrations in a femtosecond time scale by using optical pump-and-probe techniques. Such ultrafast spectroscopic techniques have already been applied for investigation of plasma oscillations. THz emission due to plasma oscillation of dense cold electron systems has been observed in modulation-doped heterojunctions[7–9] and in heavily doped bulk GaAs.[10,11] However, excitation/relaxation processes of optically excited nonequilibrium plasmons have not been clarified yet.

Recently, Kersting *et al.* proposed a new mechanism for the excitation of bulk plasmons in doped semiconductors, i.e., the screening of the surface depletion field by optically generated carriers ignites plasma oscillations of cold electrons in the doped bulk semiconductors.[10] Since this mechanism excites polarization only vertical to the surface, it is not relevant to lateral plasma oscillations observed in 2D electron systems (2DES) in heterostructures. It is therefore necessary to look for other mechanisms for excitation of lateral 2D plasma oscillations.

In this Rapid Communication, we report on THz plasmon emission from a dense cold 2DES in a modulation-doped $Al_xGa_{1-x}As/GaAs$ single quantum well (SQW) photoexcited by femtosecond laser pulses. From the dependence of the emitted THz electric field, F_{THz}, on the pump photon energy and excitation power, it has been found that there are two distinct mechanisms for 2D plasmon excitation, i.e., coherent *impulsive stimulated Raman scattering* (ISRS) and incoherent *sudden heating of 2DES*. Furthermore, by monitoring the phase of the emitted THz radiation, a clear crossover from one mechanism to the other has been observed as the pump photon energy goes through the absorption edge of the electron system.

The sample and the experimental setup used in this work were described in detail in our previous work.[8,9] The sample was a modulation-doped $Al_{0.3}Ga_{0.7}As/GaAs$ SQW grown by molecular beam epitaxy. A 170-Å-thick GaAs SQW was sandwiched by selectively doped $Al_xGa_{1-x}As$ barriers. The electron density and the low-field mobility of the sample were 9.0×10^{11} cm^{-2} and 2.6×10^5 $cm^2/V\,s$ (at 4.2 K), respectively. A NiCr/Au grating coupler with a priod (Λ) of 3 μm was fabricated on the sample surface to convert the 2D plasmon modes into radiative modes.[1]

THz emission from the SQW was measured by standard time-resolved THz emission spectroscopy.[8] The sample was cooled at $\sim$8 K. Nonequilibrium plasmon distribution was created using a mode-locked Ti:sapphire laser pulses [$\Delta\tau \sim 100$ fs; the full width at half maximum (FWHM) bandwidth $\sim$20 meV]. The laser beam was weakly focused onto the sample surface with a spot size of $\sim$350 μm at a polar angle of 45°. The THz radiation emitted along the specular beam direction was detected by a photoconductive dipole antenna gated by delayed probe laser pulses. The polarization of the incident pump beam was set to be *p*-polarized and perpendicular to the metal stripes. Polarization of the emitted THz radiation was also *p*-polarized because of the metallic grating.

Figure 1 shows traces of temporal evolution of emitted F_{THz} measured for various excitation photon energies, $\hbar\omega_L$.

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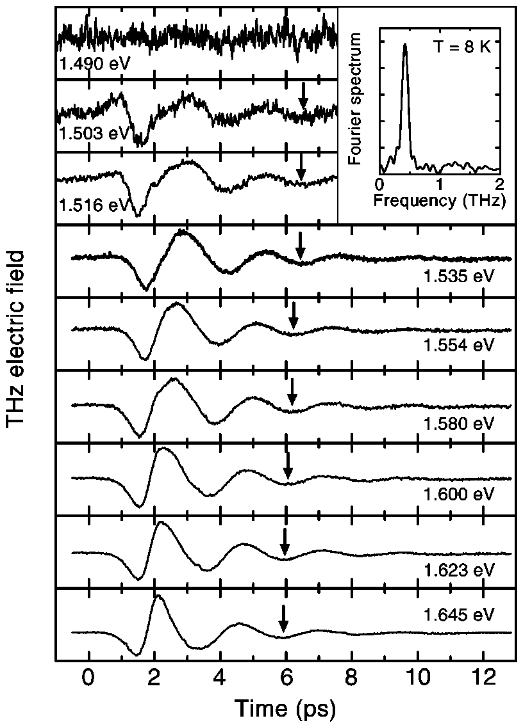

FIG. 1. Temporal evolutions of plasma oscillations measured with various pump photon energies. The inset shows a typical Fourier spectrum of the emitted THz electric field.

The power of the pump beam, P, was fixed at 100 mW. The THz transients exhibit damped sinusoidal oscillations whose Fourier spectra have a peak at ~ 0.43 THz, as shown in the inset of Fig. 1. The emission frequency is in agreement with that of the first-order plasmon mode of the 2DES predicted by a nonperturbative dispersion calculation.[12] The peak frequency did not depend on the excitation conditions within an accuracy of ± 8 GHz. The peak height of each Fourier spectrum is plotted by full circles in Fig. 2(a) as a function of $\hbar\omega_L$. It is noticed from Fig. 2(a) that the emitted THz amplitude gradually increases with increasing $\hbar\omega_L$ and exhibits a plateau at ~ 1.55 eV. A further increase of $\hbar\omega_L$ from 1.58 eV to 1.64 eV leads to an increase of F_{THz} by a factor of 2.5.

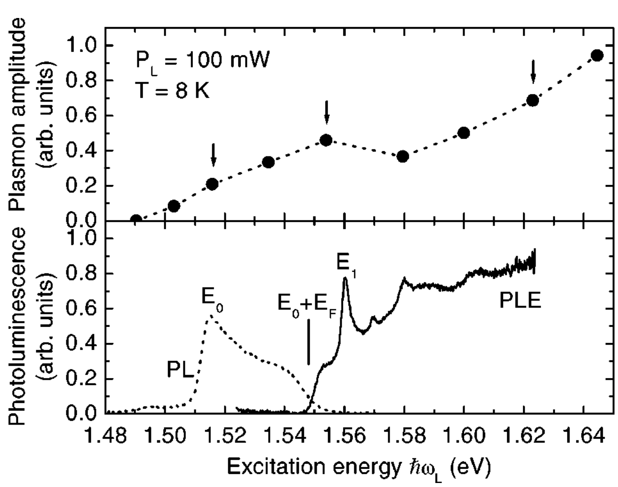

FIG. 2. (a) The dependence of the amplitude of the 2D plasma emission on the pump photon energy. (b) The photoluminescence (PL) and photoluminescence excitation (PLE) spectra are shown by a dotted and a solid line, respectively.

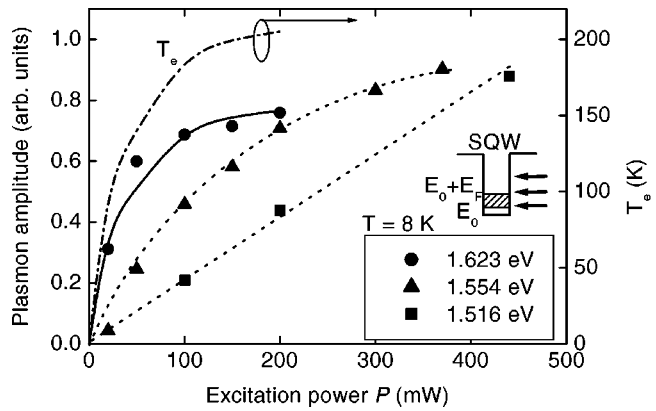

FIG. 3. Excitation power (P) dependence of the 2D plasmon amplitude at three different pump photon energies: below ($\hbar\omega_L = 1.516$ eV) and above ($\hbar\omega_L = 1.554, 1.623$ eV) the absorption edge. The solid line shows the calculated P dependence of the plasmon amplitude for $\hbar\omega_L = 1.623$ eV. The electron temperature (T_e) calculated for $\hbar\omega_L = 1.623$ eV is also plotted by a dash-dotted line. The dotted lines for 1.516 eV and 1.554 eV are guides for the eyes.

To obtain an insight into the excitation mechanism of plasma oscillations, we first investigated the correlation between the emitted THz signal and the absorption of the pump laser beam. In Fig. 2(b), the photoluminescence (PL) and photoluminescence excitation (PLE) spectra of the sample are plotted by a dotted and a solid lines, respectively. The detection energy for the PLE measurement was 1.52 eV. Reflecting a very high Fermi energy of the 2DES (~ 30 meV), the PL spectrum exhibits a broad emission band that extends from the bottom of the ground conduction subband, E_0, up to the Fermi level, $E_0 + E_F$. The shoulder structure at 1.54 eV is due to the Fermi edge singularity.[13] The PLE spectrum, on the other hand, clearly shows that the absorption takes place only for the incident light with energy above the Fermi level due to the band-filling effect. The peak seen at 1.56 eV is due to the excitonic transition to the first excited subband, E_1. Surprisingly, a relatively strong plasmon emission is observed even below the absorption edge ($\hbar\omega_L < 1.55$ eV), where the 2D electron system is transparent to the pump beam. Since the FWHM of the pump beam is 20 meV, the emission for $\hbar\omega_L < 1.55$ eV is significant.

Next, we measured the dependence of the emission amplitude on P at three different $\hbar\omega_L$'s indicated by arrows in Fig. 2(a). The obtained P dependence of the THz emission amplitudes is plotted in Fig. 3 (the inset of Fig. 3 schematically denotes the excitation energy positions). When the 2DES is pumped below the absorption edge ($\hbar\omega_L = 1.516$ eV), where the 2DES is transparent to the pump beam, the amplitude of the THz emission is found to increase linearly with increasing P. Such a linear dependence of F_{THz} on P indicates that the polarization of the photoexcited plasmons is phase-correlated with the excitation laser pulse and increases in proportion with P. When electrons are excited above the absorption edge ($\hbar\omega_L = 1.554$ eV and 1.623 eV), however, a drastic change is observed; the emitted THz electric field exhibits a sublinear dependence on P, suggesting that the phase information in the polarization is lost and incoherent processes take place in the excitation process. Such a saturation behavior is not caused by absorption saturation; the absorption saturation in QW's occurs when the photoexcited carrier density exceeds $10^{12}\ \mathrm{cm}^{-2}$,[14] while the photoexcited carrier density per pulse under the present experimental condition ($\lesssim 10^{11}\ \mathrm{cm}^{-2}$) is much lower. The difference in the P dependence can be explained by considering the following excitation mechanisms.

When the 2DES is pumped by the laser pulses whose photon energy is smaller than the absorption edge, no real electron-hole (e-h) pairs are created. However, if there exist Raman-active modes in the system, the optical field interacts with such modes via the ISRS process. Such a process can be phenomenologically expressed as[15,16]

$$\frac{d^2Q}{dt^2}+2\gamma\frac{dQ}{dt}+\omega_p^2 Q=\frac{1}{2}\left(\frac{d\chi^{(1)}}{dQ}\right)_0 F_L F_L, \tag{1}$$

where Q is an amplitude of a vibrational mode, γ the phenomenological damping constant, ω_p the eigenfrequency of the normal vibrational mode, $\chi^{(1)}$ the linear susceptibility, and F_L the electric field of the laser pulse. Since the 2D plasmon mode is Raman-active, plasma oscillations can be excited by ISRS. As seen in Eq. (1), ISRS is expected to drive the plasmon amplitude in proportion to the pump laser power $P(\propto F_L^2)$, which is the case as seen for $\hbar\omega_L = 1.516$ eV in Fig. 3. It is also noted that since the excitation pulse width ($\Delta\tau \sim 100$ fs) is much shorter than the period of the plasma oscillation ($2\pi/\omega_p$), the laser pulses exert an impulsive force on the 2DES. The effect of the impulsive nature of the photoinduced force on the phase of the plasma oscillation will be discussed later.

When the sample is pumped above the absorption edge, on the other hand, real carriers with excess kinetic energies are created. In this absorptive region, two more possible mechanisms are considered to participate in the excitation process. One is the dynamical screening of photoexcited carriers by the cold 2DES and the other is sudden heating of 2DES by ultrafast thermalization of excess energies of photoexcited carriers.

When excess electrons and holes are created by the excitation laser pulses, the cold electrons screen them. Plasma oscillations can be started by such a dynamical screening effect of 2DES. However, we think this mechanism is not relevant to the present case for the following two reasons: (1) The illumination by the pump laser pulses through the gap of the metallic grating coupler always creates a population of excess carriers whose spatial distribution is symmetric with respect to the center of the gap of the metal stripes. However, such symmetric modes are nonradiative because of the selection rule.[17,18] (2) As seen in Fig. 2, the plasmon amplitude increases by a factor of ~ 2 when $\hbar\omega_L$ is varied from 1.58 eV to 1.62 eV, while the number of photoexcited carriers does not change that much even if we take into account the increase in absorption coefficient (only $\sim 15\%$) estimated from the PLE spectrum.

Next, let us consider the other mechanism, i.e., sudden heating of 2DES. It is well established that when carriers are photogenerated above the Fermi surface of a dense cold electron system, the excess kinetic energy of photoexcited carriers is very efficiently thermalized by electron-electron scattering. A thermalization time as short as <100 fs, which is much shorter than $2\pi/\omega_p$, has been reported.[19] As a result, the excess kinetic energy is immediately converted to heat and the 2DES is suddenly warmed up. Consequently, the plasmons are thermally excited. In the thermal process, plasmon modes both symmetric and antisymmetric with respect to the grating stripes are excited and only the antisymmetric mode is selectively converted to emission.[17]

Assuming that the excess kinetic energy delivered by one laser pulse, U, is converted to heat, we have estimated the temperature, T_e, of 2DES immediately after photoexcitation by the equation $C_e(T_e-T_0)=U$, where T_0 is the equilibrium lattice temperature. C_e is the specific heat of 2DES, which is expressed as $C_e=(\pi^2/3)D_{2\mathrm{D}}(E_F)k_B^2 T_e$, where $D_{2\mathrm{D}}(E_F)$ is the density of states of 2DES at the Fermi level $(=m^*/\pi\hbar^2)$ and k_B the Boltzmann constant. From the above equations, T_e can be readily calculated as a function of the input power, P. U was estimated by taking into account reasonable values for reflection losses and the absorption coefficient of 2DES. In Fig. 3, T_e calculated for $\hbar\omega_L = 1.623$ eV is plotted as a dash-dotted line as a function of P. It should be noted that T_e is expected to exceed 100 K for the laser powers used in the present experiment.

The occupation number of plasmon modes, N_q, is then given by the Bose-Einstein distribution function, $N_q = 1/[\exp(\hbar\omega_p/k_B T_e)-1]$. This nonequilibrium plasmon distribution is the source of the THz emission. The emitted F_{THz} is expected to be proportional to $\sqrt{N_q}$. The solid line in Fig. 3 shows the calculated dependence of F_{THz} on P (the magnitude was adjusted to give the best fit to the experimental data). Although the theory is crude, the calculated dependence reasonably reproduces the experiment (circles), suggesting that sudden heating of the electron system is the dominant excitation mechanism when $\hbar\omega_L$ is well above the absorption edge.[20] This heating mechanism naturally explains the increase of F_{THz} with increasing $\hbar\omega_L$ for $\hbar\omega_L > 1.58$ eV, as seen in Fig. 2. It is known that the relaxation time, τ_ε, of the heat dumped into the electron system by the laser pulses is of the order[21,22] of 100 ps and much longer than $2\pi/\omega_p$. Therefore, the force acting on the plasmons is similar to a step (θ) function.

The difference in the nature of the force acting on the plasmons is expected to manifest itself in the time evolution of plasma oscillations. It is known that the amplitude $Q(t)$ of a harmonic oscillator is proportional to $\sin\omega t$ $(1-\cos\omega t)$ when the driving force $f(t)$ is similar to a δ function (θ function).[16,23]

Since $f(t)$ in the transparent region is considered to be δ-function-like, while $f(t)$ in the absorptive region is approximated by a θ function, a $\pi/2$ shift in the phase of the plasma oscillation is expected between the transparent and the absorptive regions. Now, let us closely look at the temporal evolutions of the plasma oscillations shown in Fig. 1.

Since F_{THz} measured immediately after the excitation is not only due to plasma oscillations but also due to other optical nonlinear effects, it is difficult to discuss the initial phase of the oscillation. Therefore, let us focus on a dip in F_{THz} observed, for example, around 6 ps (indicated by arrows in Fig. 1). It is clearly seen in the figure that the position of the dip in F_{THz} gradually shifts from 6.5 ps to 5.9 ps as $\hbar\omega_L$ is varied from 1.503 eV to 1.645 eV. Since one period of the plasma oscillation is 2.3 ps, the observed shift corresponds to a phase shift of 0.52π, which is very close to $\pi/2$. This fact strongly supports our interpretation of the excitation mechanisms.

In summary, we have systematically investigated the excitation mechanism of 2D plasmons in the selectively doped SQW by femtosecond laser pulses. From the dependence of the emitted THz electric field on the pump photon energy and excitation power, it has been found that there are two distinct mechanisms for 2D plasmon excitation, i.e., coherent *impulsive stimulated Raman scattering* and incoherent *sudden heating of 2DES*. Furthermore, by monitoring the phase of the emitted THz radiation, a clear crossover from one mechanism to the other has been observed as the pump photon energy goes through the absorption edge of the electron system.

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[1] S.J. Allen, Jr., D.C. Tsui, and R.A. Logan, Phys. Rev. Lett. **38**, 980 (1977).

[2] D. Heitmann, in *Physics and Application of Quantum Wells and Superlattices*, edited by K. von Klitzing and E. E. Mendez (Plenum, New York, 1988), p. 317.

[3] D.C. Tsui, E. Gornik, and R.A. Logan, Solid State Commun. **35**, 875 (1980).

[4] R.A. Höpfel, E. Vass, and E. Gornik, Phys. Rev. Lett. **49**, 1667 (1982).

[5] R. Höpfel *et al.*, Surf. Sci. **113**, 118 (1982).

[6] K. Hirakawa, K. Yamanaka, M. Grayson, and D.C. Tsui, Appl. Phys. Lett. **67**, 2326 (1995).

[7] K. Hirakawa *et al.*, Surf. Sci. **361**, 368 (1996).

[8] M. Voßebürger *et al.*, J. Opt. Soc. Am. B **13**, 1045 (1996).

[9] N. Sekine *et al.*, Appl. Phys. Lett. **74**, 1006 (1999).

[10] R. Kersting *et al.*, Phys. Rev. Lett. **79**, 3038 (1997).

[11] R. Kersting, J.N. Heyman, G. Strasser, and K. Unterrainer, Phys. Rev. B **58**, 4553 (1998).

[12] L. Zheng, W.L. Schaich, and A.H. MacDonald, Phys. Rev. B **41**, 8493 (1990).

[13] A. Pinczuk *et al.*, Solid State Commun. **50**, 735 (1984).

[14] D.A.B. Miller *et al.*, Appl. Phys. Lett. **41**, 679 (1982).

[15] Y.-X. Yan, E.B. Gamble, Jr., and K.A. Nelson, J. Chem. Phys. **83**, 5391 (1985).

[16] G.A. Garret, T.F. Albrecht, J.F. Whitaker, and R. Merlin, Phys. Rev. Lett. **77**, 3661 (1996).

[17] R.E. Tyson, D.E. Bangert, and H.P. Hughes, J. Appl. Phys. **76**, 5909 (1994).

[18] Strictly speaking, the selection rule is broken down for the 45° incidence. However, as calculated in Ref. 17, the coupling efficiency for the symmetric mode for the 45° incidence is only 1/400 of that of the antisymmetric mode. Therefore, the effect of the breakdown of the selection rule can be safely neglected.

[19] W.H. Knox *et al.*, Phys. Rev. Lett. **61**, 1290 (1988).

[20] Since resonant enhancement of Raman scattering is effective around the absorption edge [G. Abstreiter *et al.*, in *Light Scattering in Solids* IV, edited by M. Cardona and G. Guntherodt (Springer-Verlag, Berlin, 1984), Chap. 2, p. 45], it is expected that both stimulated Raman process and sudden heating contribute to the excitation of plasmons for the case of $\hbar\omega_L = 1.554$ eV. Indeed, as seen in Fig. 3, the plasmon amplitude for $\hbar\omega_L = 1.554$ eV shows somewhat intermediate behavior between the cases of $\hbar\omega_L = 1.516$ eV and 1.623 eV and keeps growing with increasing P even when the heating process saturates for $\hbar\omega_L = 1.623$ eV.

[21] H. Lobentanzer *et al.*, Phys. Rev. B **36**, 2954 (1987).

[22] K. Leo, W.W. Rühle, and K. Ploog, Phys. Rev. B **38**, 1947 (1988).

[23] H.J. Zeigler *et al.*, Phys. Rev. B **45**, 768 (1992).