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Time-resolved Raman scattering measurement of photoexcited plasmon in GaP crystals

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An overdamped plasmon-LO phonon coupled mode in undoped GaP crystals has been observed under high excitation condition. We have studied the relaxation process of the coupled mode in the time range of nonoseconds using a time-resolved Raman scattering technique at 81 K. The decay time of photocreated free carriers has been estimated from line-shape fitting of the coupled mode in the time-resolved Raman spectra. The decay time obtained from the Raman study has been compared with the value obtained from luminescence measurements. The plasmon hybridized with the LO phonon is found to be a gaslike electron-hole plasma.

I. INTRODUCTION

Time-resolved Raman scattering measurement provides a convenient means for investigating nonequilibrium states of molecules and solids.[1] In recent years, the time-resolved Raman scattering technique using a nanosecond pulsed laser has been applied to the study of dynamics of recrystallization process under pulsed-laser annealing.[2]

We have applied the technique to the study of photoexcited plasmon in semiconductors. Semiconductor plasmas in equilibrium state have been extensively studied by Raman scattering[3] and luminescence technique.[4] Recently, the coupled plasmon LO-phonon modes and the single particle excitation of photoexcited free carriers in GaAs have been studied by several authors.[5–8] However, the relaxation process of the coupled plasmon LO-phonon mode in photoexcited semiconductors has not been investigated systematically.

A time-resolved Raman spectrometer has been constructed in our laboratory, which consists of two nanosecond pulsed dye lasers pumped with an excimer laser. Using this apparatus, we have measured time-resolved Raman scattering spectra of highly photoexcited GaP crystals. The GaP crystal was chosen for the following reasons: (1) Raman signals from GaP crystals are strong compared with those of other III-V compounds for visible laser light. (2) In our previous single-beam experiment,[9] electronic Raman bands arising from intervalence band transitions and an overdamped plasmon LO-phonon coupled mode have been observed under high excitation condition. The dependence of the intensity of these bands on exciting laser power has been also examined. The observations allow us to use these bands as a monitor of the excess carriers in studying the relaxation process of the photoexcited carriers. (3) Since GaP is an indirect-gap semiconductor, the lifetime of excited carriers is longer than that of direct-gap semiconductors. The decay time of the electron-hole plasma deduced from luminescence measurements of GaP was found to be several tens of nanoseconds.[10] These results indicate that the GaP crystal is a suitable material for studying dynamics of excited states with a time-resolved Raman scattering technique using nanosecond pulsed dye lasers. The indirect and direct energy gaps of GaP at liquid-N_2 temperature are about $E_g = 2.3$ eV and $E_0 = 2.86$ eV, respectively. Setting the wavelengths of the probe and exciting lasers in the range between the E_0 gap and the E_g gap, we can avoid the luminescence in Raman measurements.

Using a theoretical Raman cross section, we have calculated the Raman line shape of the plasmon-LO phonon coupled mode in photoexcited GaP crystals. From the line-shape fitting of the observed plasmon-LO phonon coupled mode, we have estimated the life time of the photoexcited plasmon which couples with the LO phonon.

II. EXPERIMENTAL PROCEDURE

The experimental setup for the measurement of the time-resolved Raman scattering is shown in Fig. 1. This system consists of two pulsed dye lasers and two optical delay lines. A XeCl excimer laser operating at 40 Hz was used to excite the two dye lasers simultaneously at different wavelengths. The first dye-laser with a maximum peak power of 54 kW was used to excite GaP crystals. The second dye laser was used as a probe laser, whose peak power was about 9 kW. The pulse width of these dye lasers was about 12 ns and the spectral line width of the probe laser was about 0.8 Å. The background emission of the dye lasers was filtered with a combination of an aperture, and a grating (1200 gratings/mm) or a dispersion prism. The probe pulse was optically delayed with three 1.5-m confocal spherical mirrors, which were coated with multilayer dielectric material. The delay time was varied by changing the angle of a mirror and thereby the number of the reflection. The delay time was evaluated from counting the number of spots on the spherical mirror surface. A right-angle prism sliding on an optical bench was used for the delay of the exciting laser in order to obtain the delay times with intervals of 10 ns. Combining these optical delays, the delay times were changed with intervals of 10 ns in the range from 0 to 150 ns.

The use of two dye lasers enables us to independently excite probe and exciting lasers at different wavelengths. If the two wavelengths are separated enough, the exciting laser does not perturb the Raman measurement with the probe laser. In this experiment, we set the wavelength at 4800 Å and about 4500 Å for exciting and probe lasers, respectively. This selection of the wavelengths is necessary to prevent the Raman measurement from being obscured by intense lumi-

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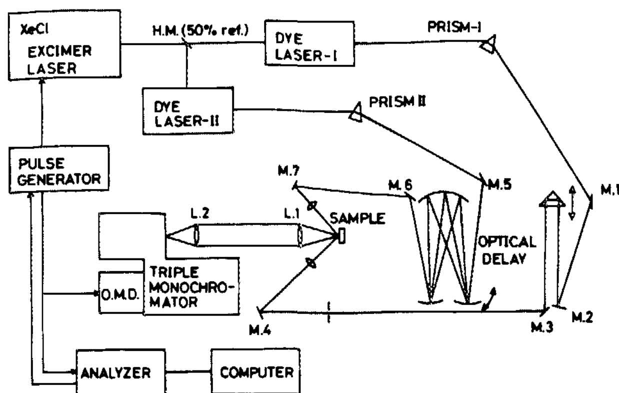

FIG. 1. Block diagram of the experimental set up for time-resolved Raman measurement.

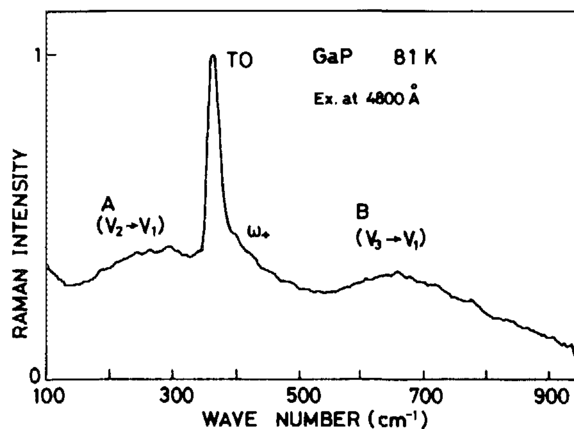

FIG. 2. The Raman spectra of GaP obtained at the highest excitation condition. The peak power of the exciting laser was about 54 kW.

nescence. The absorption coefficient of the GaP crystal at liquid-N_2 temperature at 4500 Å is 1400 cm^{-1} and that at 4800 Å is 300 cm^{-1}.[11] These wavelengths lie between the E_g and E_0 gaps of GaP crystals. The probe- and exciting-laser beams were focused on the sample surface with a cylindrical lens. The exciting laser was focused onto the sample as a line image of approximately 0.2 mm$\times$2 mm and the probe laser was focused onto the sample as a line image of approximately 0.15 mm$\times$2 mm. The position of the images is adjusted such that the probe-laser image lies in the middle of the exciting-laser image. The position of the two beams was checked with a periscope placed behind an entrance slit of a spectrometer. The intensity of the excitation laser beam is not uniform spatially. However, with this configuration the Raman signals arising from the region, which was illuminated by the exciting laser with weak intensity, were not sampled. The Raman spectra were analyzed with a triple spectrograph (SPEX 1877). A multichannel detector (Tracor-Northern TN-6133) was used to integrate the Raman signal. The spectrometer with a 1200 gratings/mm grating provides a band pass of 0.35 Å per 25 μm at the surface of the optical multichannel detector. The integration time was set to be 400 s. The acquired data were sent to a multichannel analyzer and stored in a microcomputer. The samples used in this experiment were undoped epitaxial layers of GaP crystals with (111) face grown by liquid-phase epitaxy. The electron concentration is $8.2\times10^{16}/cm^3$ at room temperature. The sample was glued with silver paste to a copper cold finger of a conduction-type liquid-N_2 cryostat. The temperature of the sample surface was about 81 K when the sample was not irradiated with the lasers. The measurements were performed with a quasibackscattering geometry.

III. RESULTS AND ANALYSIS

A. Excitation power dependence of spectra

Figure 2 shows the Raman spectra generated only with the exciting laser under high excitation condition. The exciting-laser power is 54 kW. In this spectrum two broad bands labeled "A" and "B" are observed at about 300 and 700 cm^{-1}, respectively. The intensity of these bands increases with increasing the power of the exciting laser. The bands A and B were assigned to electronic transitions from the light-hole band to the heavy-hole band ($V_2\rightarrow V_1$) and from the split-off band to the heavy-hole band ($V_3\rightarrow V_1$), respectively. The LO-phonon band at about 410 cm^{-1} is not observed in this case. The absence of the LO-phonon band was attributed to a coupling between the LO phonon and overdamped plasmon.[9] The intensity of the A and B bands was weaker than that observed under highest excitation condition of the previous single-beam experiment,[9] because in this case the excimer laser beam was divided into two beam lines to excite two dye lasers. Since the line shape of the LO-phonon band depends on the exciting-laser power to be stronger than that of the A and B bands, we monitored the line-shape of the LO-phonon band as a function of the delay time in this experiment.

We measured the dependence of Raman spectra on exciting-laser intensity when the crystals were irradiated with the exciting and probe pulses at the same time. Figure 3 shows the Raman scattering spectra obtained for several exciting-laser powers obtained at 81 K. Each spectrum is normalized with the peak intensity of the TO-phonon band. The

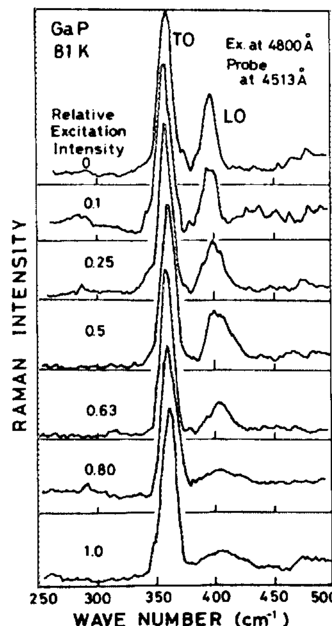

FIG. 3. The Raman spectra for various exciting-laser power at 81 K. The crystals were irradiated with the exciting- and probe-laser pulses at the same time. Each spectrum is normalized with the peak intensity of the TO-phonon band.

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spectrum at the top of this figure is measured with the probe laser only. The intensity of the probe laser is attenuated as much as possible within the range in which the Raman spectra can be observed with good signal to noise ratio. The TO- and LO-phonon bands were observed clearly. However, the intensity ratio of the TO and LO bands indicates that the high excitation effect still remains on the LO-phonon band. As the exciting-laser power is increased, the LO-phonon band broadens and almost disappears at higher excitation levels. Since the width and peak position of the TO-phonon band do not depend on the excitation level, the sample heating is ruled out for the explanation of the behavior of the LO-phonon band. Furthermore, a luminescence band of electron-hole plasma is observed at higher excitation levels. For these reasons, we attribute the broadening and disappearance of the LO-phonon band at the highest excitation level to hybridization with the overdamped plasmon, as speculated in a previous single-beam study.[9]

B. Plasmon-LO phonon coupled modes

In order to estimate the density of photocreated free carriers, a calculated line shape is fitted to the observed plasmon-LO phonon band. In general, the Raman scattering from the plasmon-LO phonon coupled modes in polar semiconductors occurs by means of three mechanisms[3]: (1) the deformation potential mechanism, (2) the electro-optic mechanism, and (3) the charge-density fluctuation mechanism. The LO-overdamped plasmon mode in doped *n*-GaP has been examined by Hon and Faust[12] and Irmer *et al.*[13] The Raman efficiency given by Irmer *et al.* consists of A and B terms. The A term represents the Raman efficiency due to the mechanisms (1) and (2). The B term represents the Raman efficiency due to the mechanism (3). Hon and Faust have taken into account only the mechanisms (1) and (2). The expression of Raman efficiency given by Hon and Faust[12] is equivalent to the A term of that given by Irmer *et al.*[13] These authors have suggested that Raman scattering from coupled modes in *n*-GaP occurs mainly by the deformation-potential and electro-optic mechanisms. The contribution of the charge-density fluctuation mechanism to the Raman efficiency diminishes at the LO-phonon frequency[14] and moreover distorts the line shape of the coupled mode. However, our observation indicated that the Raman scattering intensity was not zero at the LO-phonon frequency. Accordingly, the contribution of the charge-density fluctuation mechanism to the Raman efficiency of the coupled mode may be small in photoexcited GaP. Combining these suggestions and our experimental results, we assume that the electro-optic and deformation-potential mechanisms are dominant for the Raman scattering from the plasmon-LO phonon coupled mode in photoexcited GaP crystals as well as in doped GaP crystals. We employ the Raman efficiency given by Hon and Faust,[12] which is expressed by

$$\frac{\partial^2\sigma}{\partial\Omega\partial\omega} = \mathrm{Im}\{[-\epsilon_\infty/\epsilon(\omega)][1+2CL(\omega)-C^2 \times(\omega_T^2/\Omega^2)(1+\omega_p^2 P(\omega))L(\omega)]\} \times(4\pi/\epsilon_\infty)(n_\omega+1), \quad (1)$$

$$L(\omega) = [1-(\omega^2/\omega_T^2)-(i\omega\Gamma/\omega_T^2)]^{-1}, \quad (2)$$

$$P(\omega) = -(\omega^2+i\omega\gamma)^{-1}, \quad (3)$$

$$\epsilon(\omega) = \epsilon_\infty[1+\omega_p^2 P(\omega)+(\Omega^2/\omega_T^2)L(\omega)], \quad (4)$$

$$\omega_p^2 = 4\pi ne^2/m^*\epsilon_\infty, \quad (5)$$

γ = plasmon damping,
Γ = phonon damping,
ω_T = TO-phonon frequency,
ω_L = LO-phonon frequency,
C = Faust–Henry coefficient,
ω_p = plasma frequency,
n = free carrier density,

and Ω is related with ω_L and ω_T via

$$\omega_L^2 = \omega_T^2+\Omega^2. \quad (6)$$

The interference between the deformation-potential mechanism and the electro-optic mechanism is a remarkable feature of this model. The degree of the interference depends on the Faust–Henry coefficient[15] ($C = -0.53$).[12] Photocreated plasmons consist of two types of mobile carriers. Using the Hall mobilities of the electron[16] ($\mu_e = 700$ cm^2/V s) and hole[17] ($\mu_h = 360$ cm^2/V s) and the effective masses of the electron[18] ($m_e^* = 0.35m_0$) and hole[19] ($m_h^* = 0.41m_0$) at about 80 K, we have estimated the electron- and hole-damping constants. These effective masses are defined as follows:

$$\frac{1}{m_e^*} = \frac{1}{3}\left(\frac{2}{m_t^*}+\frac{1}{m_l^*}\right), \quad (7)$$

$$\frac{1}{m_h^*} = \frac{1}{m_{\mathrm{lh}}^*}\frac{1}{1+(m_{\mathrm{hh}}^*/m_{\mathrm{lh}}^*)^{3/2}} + \frac{1}{m_{\mathrm{hh}}^*}\frac{1}{1+(m_{\mathrm{lh}}^*/m_{\mathrm{hh}}^*)^{3/2}}, \quad (8)$$

where

$m_t^* = 0.254m_0$, $m_l^* = 1.7m_0$,
$m_{\mathrm{hh}}^* = 0.55m_0$, and $m_{\mathrm{lh}}^* = 0.13m_0$.

The Hall mobility μ and the carrier damping γ are related with the following equation;

$$\gamma = |e|/m^*\mu. \quad (9)$$

The electron- and hole-damping constants of GaP calculated using Eq. (9) were $\gamma_e = 38$ cm^{-1} and $\gamma_h = 42$ cm^{-1}, respectively. The optic-plasma frequency was given by the following forms:

$$\omega_p^2 = \omega_{pe}^2+\omega_{ph}^2, \quad (10)$$

$$\omega_{pi}^2 = 4\pi e^2 n_i/\epsilon_\infty m_i^* \quad (i=e,h). \quad (11)$$

The density of free electrons is nearly the same as that of free holes under the high excitation condition. Then, Eq. (10) is rewritten as

$$\omega_p^2 = \frac{4\pi ne^2}{\epsilon_\infty}\left(\frac{1}{m_e^*}+\frac{1}{m_h^*}\right). \quad (12)$$

Since the damping of free electrons is considered to be nearly equal to that of free holes, the contributions of electron and hole to the dielectric function are related to the ratio of the electron- and hole-effective masses. For this reason, we have

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used the effective mass of the electron-hole plasma ($m_{ehp} = 0.18\, m_0$) as the mass of the carriers in the calculation. In recent work, the large damping of the coupled mode in photoexcited GaAs has been explained in terms of the contribution of the intervalence band transitions of the photocreated holes to the dielectric function.[5] However, we neglect the contribution of intervalence band transitions of photocreated holes to the dielectric function.

Using the plasmon frequency and damping constant as adjustable parameters, we have performed the line-shape fitting of calculated coupled mode bands to the observed ones. Other parameters are fixed as $\omega_T = 368$ cm^{-1}, $\omega_L = 407$ cm^{-1}, $\Gamma = 2$ cm^{-1}, $C = -0.53$, and $\Omega = 173.9$ cm^{-1}.

Figures 4(a) and 4(b) show the excitation power dependence of photoexcited carrier density and damping constant estimated from the Raman line-shape fitting, respectively. As the exciting-laser power is increased, the carrier density increases linearly, as shown in Fig. 4(a). The estimated free-carrier densities at the highest exciting-laser power level and without exciting laser were 3.2 and 1.4×10^{18} cm^{-3}, respectively. Since the unirradiated GaP crystal contains carriers with a concentration of $N_0 = 8.2 \times 10^{16}$ cm^{-3}, the density of photoexcited excess carriers, which are created with the probe laser, is about 1.3×10^{18} cm^{-3}. The value of the plasmon damping constant increases linearly from 1200 to 1400 cm^{-1} with increasing exciting-laser power. Since Hall measurements indicated that the damping of free carriers was about 40 cm^{-1} for undoped crystals,[16,17] it is evident that the photoexcited plasmon suffers larger damping than the plasmon in doped GaP. The reasons of the strong damping of the coupled mode in photoexcited GaP are not clear at present. A plausible explanation is as follows: The electronic Raman band due to the intervalence band transitions ($V_2 \rightarrow V_1$) of photocreated holes has been observed in GaP.[9] This electronic Raman band overlaps the LO-plasmon coupled mode. Hence, the LO-phonon photoexcited plasmon system will be heavily damped by the energy decay via the intervalence band transitions of photocreated holes. It has been also suggested that the intervalence band transitions of photocreated holes have a substantial influence on the damping of the excited plasmons.[20,21] Recent Raman measurements on photoexcited semiconductors have shown that the damping of carriers is large compared with that of carriers in thermal equilibrium.[5,6,8]

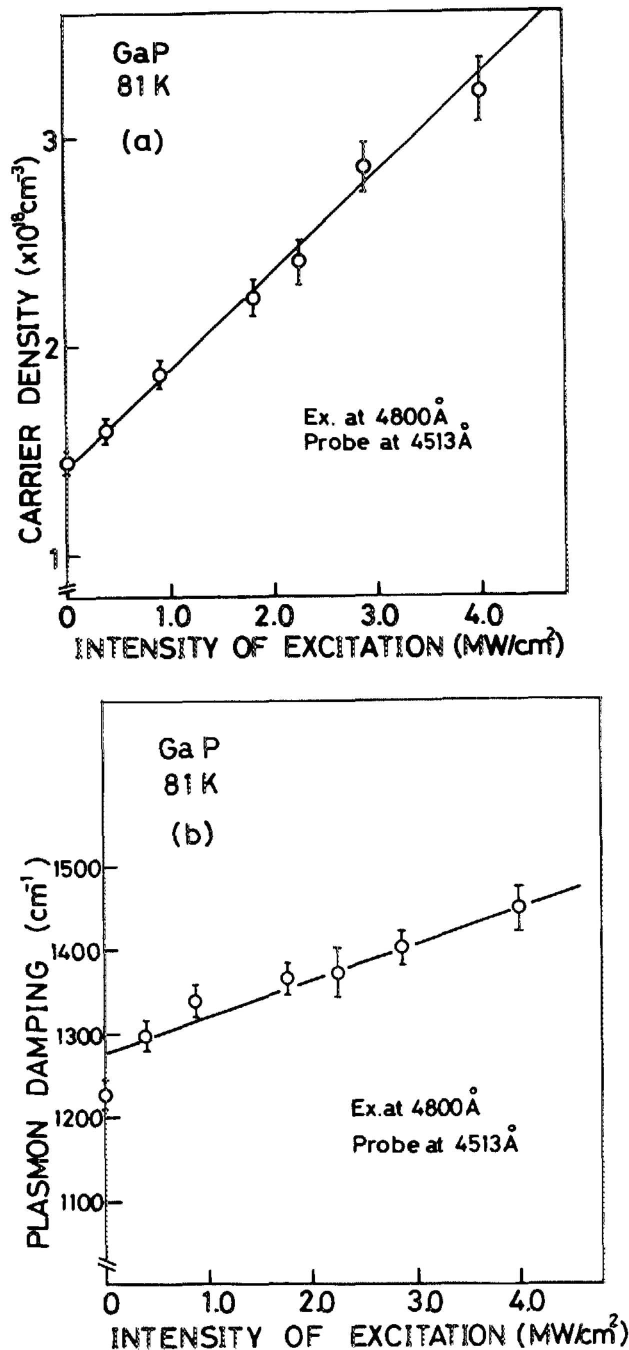

FIG. 4. The excitation power dependence of the density (a) and damping constant (b) of photocreated carriers obtained by the present analysis.

C. Time-resolved Raman spectra

Figure 5 shows the Raman spectra measured as a function of the delay time. Each spectrum is normalized with the intensity of the TO-phonon peak intensity. The spectrum at the top of this figure is measured without the excitation laser pulse, i.e., the probe pulse only. In this spectrum both TO-

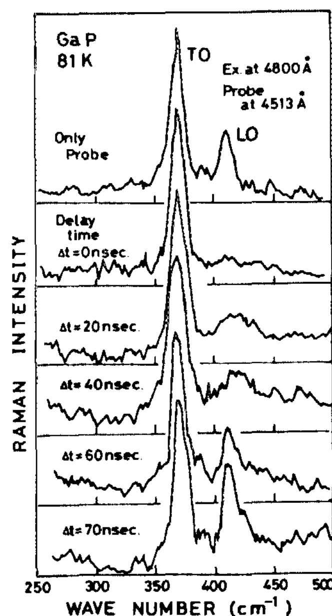

FIG. 5. The Raman spectra of GaP for various delay time. The spectrum at the top of this figure is measured with the probe laser only. Each spectrum is normalized with the peak intensity of the TO-phonon band.

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and LO-phonon bands are observed clearly. Irradiating the sample with the excitation laser without delay times, the LO-phonon band disappears completely owing to the hybridization with the overdamped plasmon. The intensity of the LO-phonon band recovers gradually with increasing delay time. This corresponds to the decrease of the free-carrier density. From the line-shape fitting of the calculated coupled mode bands to observed ones using Eqs. (1)–(5), we have estimated the free-carrier densities at various delay times. The free-carrier density is plotted against several delay times in Fig. 6. As seen in this figure, the density of the photocreated free carriers decreases with the delay time. We have assumed that the free-carrier density D exhibits a single exponential decay with time, as shown in inset of Fig. 6. The background carrier density (1.29×10^{18} cm^{-3}) corresponds to the photoexcited one created with the probe laser. The best-fit curve is represented by the solid line in this figure. From this fitting, it is determined that the free-carrier lifetime is 38 ns.

Luminescence spectra of electron-hole drop (EHD) in highly excited GaP have been studied by many workers[10,22–24] and the reported decay times of EHD are 28.5 ns,[10] 35 ns,[24] and 38 ns (Ref. 22) at 2 K. As temperature increases, the decay time of the EHD becomes shorter than that observed at 2 K.[10] The luminescence spectra of the EHD have been measured by Schwabe *et al.*[10] in highly excited GaP at 77 K. They have observed a broad luminescence band at 2.268 eV at $T = 77$ K under higher excitation condition ($I_{max} = 5$ MW/cm^2). From the excitation power dependence of this luminescence band, they have concluded that the luminescence band at $T = 77$ K consists of the EHD luminescence and the gaslike electron-hole plasma (EHP) luminescence. The EHD luminescence and gaslike EHP luminescence dominated in the low- and high-energy sides of the band, respectively. They have reported that the decay time of the gaslike EHP luminescence is 49 ns and the value of the EHD luminescence is about 25 ns at 77 K.[10] The decay time estimated from the present Raman study is somewhat different from the value of the EHD decay time but is rather close to the value of the decay time for the gaslike electron-hole plasma at 77 K. From this result, it is more likely that the electron-hole system which interacts with the LO phonon is the gaslike EHP at 81 K.

In order to support this speculation, we have measured luminescence spectra of the GaP crystal. Figure 7 shows luminescence spectra of the GaP crystal observed at several exciting laser powers at 81 K. At low excitation levels, a donor (S)–bound-exciton line is observed at about 2.3 eV. The intensity of a broad luminescence band centered at 2.285 eV increases as the exciting-laser power was increased. Under the highest excitation condition, the luminescence band centered at 2.285 eV with a half-width of about 55 meV is dominant. This broad luminescence band has a typical electron-hole plasma line shape. Further, the peak position and the half-width of the luminescence band are in good agreement with the experimental results obtained by Schwabe *et al.*[10] at 77 K under slightly low excitation levels and by Schweizer *et al.*[23] at 87 K. These authors have ascribed the luminescence band centered at 2.285 eV to a radiative recombination from the electron-hole plasma (EHP). These results indicate that the EHP state is stable in GaP crystals at 81 K under high excitation condition and thereby lend support to the conclusion that the electron-hole system which interacts with the LO phonon is the gaslike EHP.

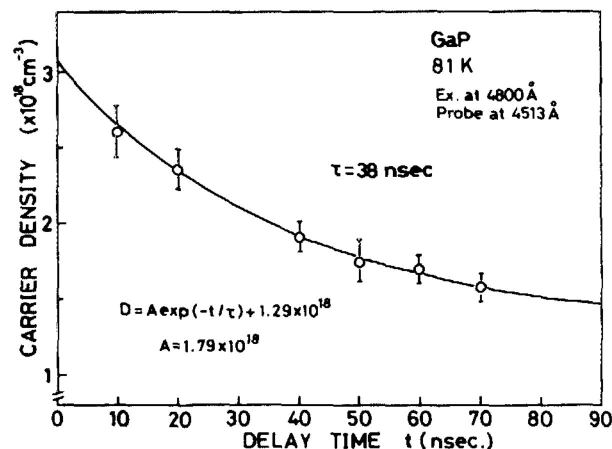

FIG. 6. Carrier density of photoexcited GaP as a function of delay time. The open circles correspond to the carrier density obtained in the present analysis. The solid line represents the calculated decay curve of photocreated carriers using the equation shown in the inset.

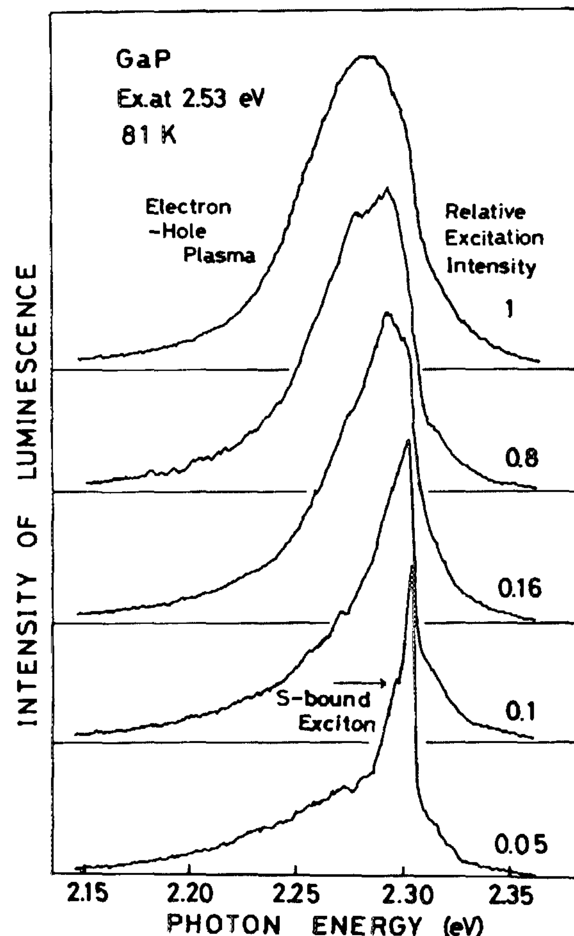

FIG. 7. The luminescence spectra of GaP for several exciting-laser powers. Each spectrum is normalized to unity with the peak intensity.

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IV. CONCLUSION

We have measured time-resolved Raman spectra of highly excited GaP crystals. From the analysis of the time-resolved Raman spectra, we have determined the decay time of photocreated free carriers ($\tau = 38$ ns). The decay time obtained in this study is close to the one obtained from the luminescence measurements of the electron-hole plasma. This result leads to the conclusion that in GaP the LO phonon hybridizes with the gaslike electron-hole plasma above 70 K. The present study demonstrates that time-resolved Raman measurements using two laser beams with different wavelengths give direct information on the relaxation process of electronic excited states of semiconductors.

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