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Raman Scattering Measurement of the Free-Carrier Concentration in $Al_xGa_{1-x}As$

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Raman scattering from coupled plasmon-optical phonon modes has been studied in direct bandgap n-$Al_xGa_{1-x}As$ layers in order to determine the carrier concentrations in the epitaxied layers. The dielectric constant method can be used to analyze the behavior of the coupled modes. This analytic treatment exhibits three new normal modes due to the coupling of plasmons and phonons, in which the mode frequencies change with the carrier concentration. The theoretical coupled mode frequencies are in good agreement with the observed frequencies in Raman scattering spectra of differently doped n-$Al_xGa_{1-x}As$ layers grown by molecular beam epitaxy.

§1. Introduction

In polar semiconductors with zinc blende-type structure, such as GaAs, InP, and GaSb, the conduction-electron plasmons couple to the longitudinal-optical(LO) phonons through the net electric dipole moment due to the lack of inversion symmetry. The coupled plasmon-LO phonon collective modes participate in the first-order Raman scattering.

Raman scattering by the coupled plasmon-LO phonon modes in GaAs has been reported by many researchers[1,2] Their spectra, which were obtained for the samples with different carrier concentrations, clearly show two Raman lines due to the coupled modes. The frequencies of the two lines change as a function of carrier concentration. Thus, the coupled mode frequency can be used to estimate the carrier concentration of the polar semiconductors. However, no detailed study has been reported for the plasmon-phonon coupling in mixed crystals.

The conduction-electrons in $Al_xGa_{1-x}As$ with direct bandgap are expected to couple to the LO phonons by the macroscopic electric field, since the small effective mass of the electrons in the Γ conduction band produces the high-density electron gas. In order to study the coupling modes of mixed crystals, it is required to use the samples with homogeneous composition and uniform carrier concentration. In this report, we describe Raman scattering from the coupled plasmon-LO phonon modes in direct bandgap $Al_xGa_{1-x}As$ grown by molecular beam epitaxy (MBE) and its application for the determination of the carrier concentration. This method has great advantages for determining simultaneously the alloy composition and the carrier concentration in a nondestructive way at room temperature.

§2. Theory

The frequencies of the coupled modes are the roots of the equation when the total dielectric constant for the plasmon-phonon combined system is zero[1] This dielectric constant method has been already performed on analysis of the modes in GaAs[1,3] The behavior of the coupled modes in $Al_xGa_{1-x}As$, however, is different from that in GaAs since $Al_xGa_{1-x}As$ exhibits separate GaAs-like and AlAs-like optical phonon branches. Assuming negligible phonon and electron dampings, the scattering wavevector and frequency dependent dielectric constant[3] $\varepsilon_T(q,\omega)$ in the long-wavelength limit for the combined system in $Al_xGa_{1-x}As$ can be written as

$$\varepsilon_T(q,\omega)=\varepsilon_\infty+\sum_{j=1}^{2} S_j\Big/\left\{1-(\omega/\omega_{tj})^2\right\}-\varepsilon_\infty\omega_p^2(q)/\omega^2 \quad (1)$$

where j=1, 2 refers to GaAs, AlAs, respectively. ε_∞ is the high-frequency dielectric constant, ω_{tj} is the transverse optical (TO) phonon frequency, S_j is the j-th oscillator strength, $\omega_p(q)$ is the wavevector dependent plasma frequency.

$$\omega_p^2(q) = \omega_p^2 + 0.6\,(qV_F)^2 \qquad (2)$$

and

$$\omega_p^2 = 4\pi n e^2/\varepsilon_\infty m^* \qquad (3)$$

where q is the scattering wavevector, n is the carrier concentration, V_F is the Fermi velocity, and m^* is the effective mass of the carriers. The coupled mode frequencies ω_Z are derived by equating the total dielectric constant ε_T to zero. The oscillator strengths S_1 and S_2 are obtained as the solutions of the following equations

$$S_1 + S_2 = \varepsilon_\infty \{(\omega_{\ell 1}\omega_{\ell 2}/\omega_{t1}\omega_{t2})^2 - 1\} \qquad (4)$$

$$(1+S_1/\varepsilon_\infty)\omega_{t1}^2 + (1+S_2/\varepsilon_\infty)\omega_{t2}^2 = \omega_{\ell 1}^2 + \omega_{\ell 2}^2 \qquad (5)$$

where $\omega_{\ell j}$ is the LO phonon frequency. The optical phonon frequencies $\omega_{\ell j}$, ω_{tj} and ε_∞ change with the alloy composition. Therefore, the coupled mode frequencies ω_Z of $Al_xGa_{1-x}As$ depend on both n and the Al content x.

Figure 1 shows an example of theoretically calculated curves for n dependence of ω_Z in n-$Al_xGa_{1-x}As$(x=0.3). As shown in the figure, there exist three new normal modes (denoted L+, Lo and L_) in the phonon-plasmon system, while only two coupled modes are allowed in degenerate GaAs. In Fig. 1, when n increases, the high-frequency mode L+ shifts from the AlAs-like LO phonon position to higher frequency side, the low-frequency mode L_ approaches the GaAs-like TO phonon position, and the intermediate-frequency mode Lo shifts from the GaAs-like LO phonon position to the AlAs-like TO phonon position. Thus, the coupled mode frequency can be used to determine the carrier concentration of direct bandgap n-$Al_xGa_{1-x}As$.

§3. Experimental procedures

3-1 Sample preparations

The $Al_xGa_{1-x}As$ epitaxial layers were grown on Cr-doped <100> oriented GaAs substrates in a VARIAN MBE GEN /II system.

The substrate temperature during growth was ~ 730 °C. All the samples have a four-layer structure of GaAs/$Al_xGa_{1-x}As$. The first layer grown on the substrate is a ~1 µm thick undoped GaAs buffer layer to obtain a good crystallographic quality for succeeding layers. The next layer is a ~0.5 µm thick undoped $Al_xGa_{1-x}As$ buffer layer. This is followed by a ~3 µm thick Si doped n-$Al_xGa_{1-x}As$ layer which is used in the Raman scattering experiments. The final layer is a thin (~300 Å) GaAs layer which provides a good ohmic contact in Hall effect measurements. The final layers were etched off for the Raman scattering measurements of Si doped $Al_xGa_{1-x}As$ layers.

3-2 Raman scattering measurements

Most Raman spectra were obtained at room temperature using the 5145 Å emission line from an argon ion laser. Some data were taken with the other argon laser lines and the krypton ion laser lines. Since $Al_xGa_{1-x}As$ is opaque to these laser lines, Raman scattering experiments were carried out in backscattering geometry. The selection rules for this geometry allow only the first scattering by LO phonons.

The scattered light was collected by a photographic optics and focused onto the slit of a JOBIN - YVON RAMANOR U - 1000 double holographic grating monochromator. The frequencies were calibrated using the Rayleigh and plasma lines in the lasers. The slit widths commonly used gave resolution of ~4 cm^{-1}. A cooled Hamamatsu R-649 photomultiplier with associated photon counting electronics detection was employed. The data were recorded on an X-Y plotter.

§4. Results and Discussion

In Fig. 2 the typical Raman spectra at room temperature from the (100)surfaces of $Al_{0.19}Ga_{0.81}As$ layers are shown for different carrier concentrations of 1.7 x 10^{17}, 7.1 x 10^{17}, 8.8 x 10^{17}, and 3.7 x 10^{18}cm^{-3}. The carrier concentrations in the figure are the results from the Hall effect measurements. Only two LO (GaAs-like and AlAs-like) phonons are observed in the sample with lower carrier concentration n~1.7 x 10^{17}cm^{-3}. When n increases to 7.1 x 10^{17}cm^{-3}, two shoulders (L+, Lo) appear on the high-frequency side of each LO phonon, and a broader mode (L_) is also observed on the low-frequency side of the GaAs-like LO phonon. The all compositions are determined by the frequencies of the two LO phonons originating from the surface depletion layer. For the sample with n~8.8 x 10^{17}cm^{-3}, the L+ and Lo modes are separated clearly from each LO phonon peak, and the L_ mode sharpens. At the highest carrier concentration n ~ 3.7 x 10^{18}cm^{-3}, the L- and Lo modes vibrate at the GaAs-like and the AlAs-like TO phonon frequency, respectively, while the L+ mode shifts to still higher frequency.

Equations (1) and (2) indicate the wavevector dependence of the coupled mode frequencies. Actually, it has been demonstrated that the scattering wavevector involved in Raman scattering experiments is large enough to observe wavevector dependent plasmon-LO phonon modes of degenerate n-GaAs.[4-6] Three Raman lines (L+, Lo and L_) from n-$Al_xGa_{1-x}As$ were also assigned to the coupled modes by the wavevector dependence using different laser lines.

The experimental results in Fig. 2 are compared with the numerically calculated values. The solid curves in Fig. 3 show the calculated coupled mode frequencies in n-$Al_{0.19}Ga_{0.81}As$. The observed coupled mode frequencies of each sample are plotted as open circles in Fig. 3. It is found in Fig. 3 that the experimental results agree well with the theoretical curves. Accordingly, the curves shown in Fig. 3 can be used to determine the carrier concentration of $Al_{0.19}Ga_{0.81}As$.

The L+ mode frequencies give accurate values of the high carrier concentration ($n \gtrsim 5 \times 10^{17}$ cm^{-3}) of the layers because the L+ mode frequency shifts intensively to higher energies with increasing the carrier cencentration in this range, as shown in Figs.1 and 3. On the other hand, the L_- modes are useful for the carrier concentrations of the lightly doped ($\sim 1 \times 10^{16} \leq n \lesssim 5 \times 10^{17} cm^{-3}$) layers because the frequency shifts strongly to lower energies with decreasing the carrier concentration in the range.

This measurement by Raman scattering at room temperature can be easily applied for all other n-$Al_xGa_{1-x}As$ layers in the direct bandgap region.

§5. Conclusion

We have shown that Raman scattering by plasmon-LO phonon coupled modes is a convenient way of measuring the free-carrier concentration in direct bandgap n-$Al_xGa_{1-x}As$. Raman results indicate that the coupling between LO phonons and plasmons in degenerate $Al_xGa_{1-x}As$ produces the three normal vibrational modes, as predicted from a theoretical analysis using the dielectric constant method. The coupled mode frequencies for any carrier concentration depend on the alloy composition because the composition variation changes the optical phonon frequencies, the effective mass of the carriers, and the high-frequency dielectric constant value. The observed shifts of the coupling mode frequencies in MBE n-$Al_xGa_{1-x}As$ with different doping levles fit the numercially calculated results of carrier concentration dependence.

The advantages of this Raman method are that no contacts for sample preparations, neither cooling nor heating for the measurements are necessary, and that the carrier concentration can be determined within very small areas by focusing the laser beam. In addition, the alloy composition can be also determined by measuring the frequencies of GaAs-like and AlAs-like LO phonons from the surface depletion layer. This work can be also extended to other mixed crystals.

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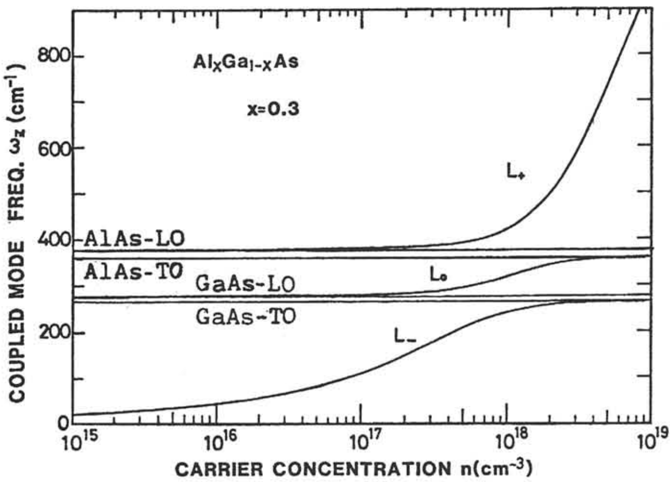

Fig. 1 Calculated coupled mode frequencies ω_Z vs. the carrier concentration n of $Al_{0.3}Ga_{0.7}As$. GaAs-like and GaAs-like optical phonon frequencies are shown as the straight lines.

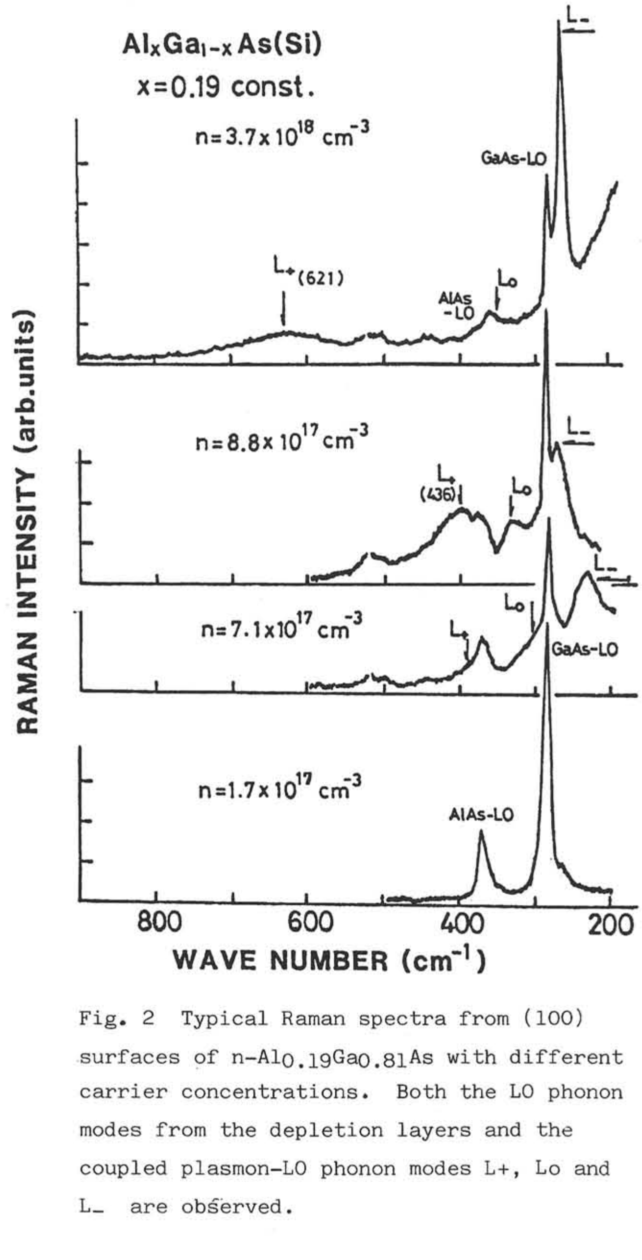

Fig. 2 Typical Raman spectra from (100) surfaces of $n\text{-}Al_{0.19}Ga_{0.81}As$ with different carrier concentrations. Both the LO phonon modes from the depletion layers and the coupled plasmon-LO phonon modes L+, Lo and L_- are observed.

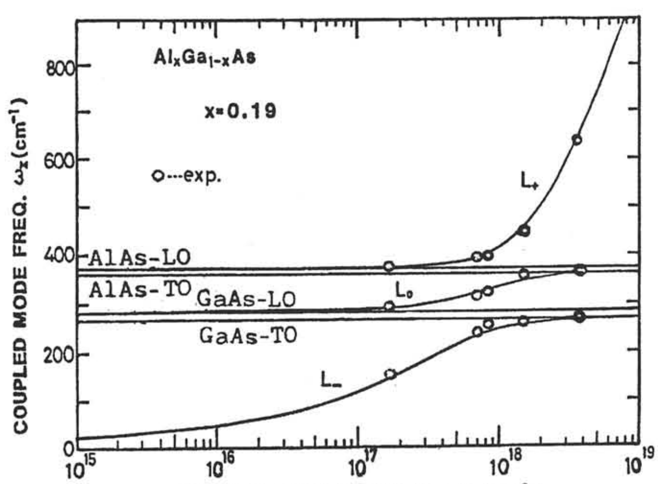

Fig. 3 The dependence of the coupled mode frequencies ω_Z on the carrier concentration n in $n\text{-}Al_{0.19}Ga_{0.81}As$. The solid lines are the calculated curves. The experimental open circles are results from Fig. 2