Interference of 2 LO phonon and continuum inter-valence band transition in p-GaInP film

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***Abstract*—Quantum interference between two LO phonon modes and valence band states is investigated by Raman spectra of p-type $Ga_{0.5}In_{0.5}P$. The spectrum fitting analysis reveals the quantum interference features in asymmetric line shape, broadening, and peak shift. This finding suggests the possibility of the control of the absorption profile in THz frequency region.**

I. INTRODUCTION

Asymmetric line shape based on quantum interference between discrete states and a continuum was theoretically presented by Fano [1]. Harris proposed a mechanics of laser without population inversion based on the quantum interference of two discrete states and a continuum, which is known as electromagnetic induced transparency (EIT) [2]. EIT was studied on the atomic system at first [3]. The studies on solid states, in particular, semiconductors followed that. Since the interaction energy width of phonons with radiation is generally less than 1 meV at room temperature (RT), which is much smaller than that of electron-radiation system, phonons are treated as discrete states in optical spectra. However, the studies on phonon systems are only for the interaction of one phonon mode with a continuum. In the studies of Fano interference of phonon systems, it was reported that phonons interact with valence bands via electronic inter-valence band transitions. This type of interaction was experimentally observed as asymmetric spectrum profiles in Raman measurements for p-type semiconductors [4-6]. Here, the increase in the hole density enhanced the asymmetry of the spectrum shape, LO phonon energy shift, and the peak broadening. It is expected that the study on two phonon mode systems leads to EIT and thus radiation and modulation devices in THz region. It is known that the disordered GaInP alloys have two LO modes producing the atomic oscillations in the same plane [7]. In this research, an alloy system of $Ga_{0.5}In_{0.5}P$ is focused, and Raman spectra of p-type $Ga_{0.5}In_{0.5}P$ films were investigated.

II. EXPERIMENT

$Ga_{0.5}In_{0.5}P$ films were grown on GaAs substrates with the surface orientations of (100) inclined to [011] by 10° for p-type samples and (511) for undoped sample by MOVPE. The film thickness was approximately 1μm. The p-type films were obtained by doping Zn atoms. Net acceptor density was measured by electrolyte capacitance-voltage method, and found to be in a range of $(5.4 - 18)\times10^{17}$ cm^{-3}. Raman measurements were conducted under a back scattering condition using the 785 nm line of a diode laser. The photon energy of this laser was smaller than the bandgap energy of $Ga_{0.5}In_{0.5}P$ and greater than that of GaAs. The luminescence from the GaAs substrate was eliminated by etching the substrates using a solution of H_2O_2 : H_2SO_4 : H_2O = 1: 4: 1. Figure 2 shows the experimental spectra of p-type and undoped $Ga_{0.5}In_{0.5}P$ films. The peaks at 330 cm^{-1}, 360cm^{-1} and 380 cm^{-1} were attributed to the respective modes of InP-like TO (TO_1), InP-like LO (LO_1) and GaP-like LO (LO_2), according to references. The TO modes are forbidden in principle in the backscattering geometry from a (100) plane of zinc-blende type crystals. The observation of this mode in the present measurements were attributed to the break of momentum conservation rule caused by the lattice fluctuation [8].

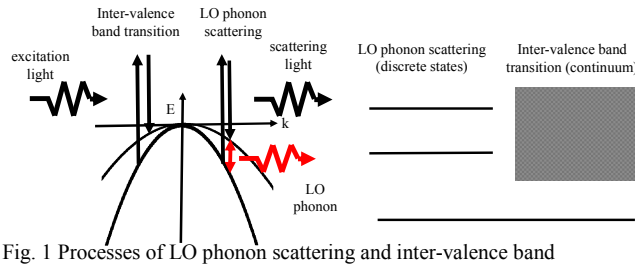

Fig. 1 Processes of LO phonon scattering and inter-valence band transition, and the energy diagram of discrete states and a continuum.

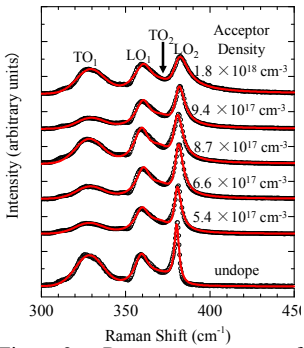

Fig. 2 Raman spectra of p-$Ga_{0.5}In_{0.5}P$ films. Solid curves show the best-fit functions.

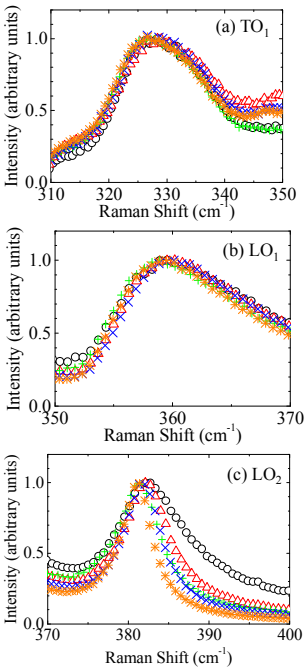

Fig. 3 Normalized Raman spectra. ○ : 18, △ : 9.4, + : 8.7, × : 6.6, ✳ :5.4 ($\times10^{17}$ cm^{-3}).

III. RESULTS

A. Qualitative analysis

Figure 3 shows the normalized Raman spectra. Here, the background due to the Raleigh scattering was subtracted. There is no change in the line shape around the TO_1 peak for the variation of the hole density. The peak of LO_2 is asymmetrically broadened in the high energy side, but the asymmetry for the LO_1 peak is smaller. It is possible that this small asymmetry is attributed to the cancellation of asymmetric spectrum features owing to the broadening of the higher energy side of the LO_1 peak and sharpening of the lower energy side of the LO_2 peak. Thus the qualitative analysis of the spectrum shape suggests the appearance of the quantum interference of the two LO modes

and a continuum in the Raman spectra. Quantitative analysis was conducted by the spectrum fitting in the following.

B. Quantitative analysis

The Raman spectra of the p-type samples were fitted by eq. (1) constructed on the basis of the Fano's and Harris's expressions.

$$f(E) = \sqrt{E}\frac{(\varepsilon_1 q_2 + \varepsilon_2 q_1 + \varepsilon_1\varepsilon_2)^2}{\varepsilon_1{}^2\varepsilon_2{}^2 + (\varepsilon_1 + \varepsilon_2)^2}, \quad (1)$$

where

$$\varepsilon_j = \frac{E - E_j - \Delta E_j}{\Gamma_j + \gamma_j} \quad (2)$$

Here, asymmetry parameter are denoted by q_j (j=1,2) for the respective LO_j modes. E_j and γ_j are the original LO_j photon energy of the mode j and its line broadening without interference, and ΔE_j and Γ_j are the energy shift and broadening due to the quantum interference. The term of $\sqrt{E}$ originates from the function of the density of states of the inter-valence band transition. The TO phonon peaks were fitted by Gaussian functions. GaP-like TO mode (TO_2) observed in a former research [9] was taken into account. Background level was approximated to be constant. In the fitting of an undoped sample, Lorentzian functions for the LO modes and Gaussian functions for the TO modes were adopted. E_j and γ_j were obtained by the fitting of an undoped sample. The solid curves in Fig. 2 are the results. It is found that the experimental spectra are well reproduced.

IV. DISCUSSION

A. Asymmetry parameter q

Figure 4 shows asymmetry parameter q_j (j=1, 2) obtained by the fitting. It is known that asymmetric feature of the line shape becomes eminent as the decrease in $|q|$, and the sign of q_j decides which side of the peak is broadened asymmetrically. The result of q_2>0 and the decreasing in $|q_2|$ as the increase in the acceptor density agrees with the feature that the LO_2 peak is asymmetrically broadened in the high energy side as shown in Fig. 3(c). The same sign of q_1 and q_2 accounts for the reduction of the asymmetric features of the spectrum shape in between 360 – 380 cm^{-1}. The decrease in $|q|$ as the increase in the hole density mostly agrees with the results on other semiconductors, for example, Si, Ge, and GaAs [4-6].

B. Energy shift ΔE

The energy shift ΔE_j from the original LO_j phonon energy was calculated by equation (3).

$$\Delta E_j = (E_{LO_j,p-type} - E_{TO_1,p-type}) - (E_{LO_j,undope} - E_{TO_1,undope}) \quad (3)$$

$E_{LO_j,p-type}$, $E_{TO_1,p-type}$, $E_{LO_j,undope}$, and $E_{TO_1,undope}$ show phonon energies obtained from the spectrum fitting on p-type and undoped samples. The results are shown in Fig. 5. The contribution of strain was removed on the basis of the TO_1 energy shift. Thus it is thought that the LO energy shift is due to the quantum interference.

C. Broadening Γ

Figure 6 shows the broadening Γ_j. Since the spectrum broadening was not observed on the peak of TO_1 mode (Fig. 3

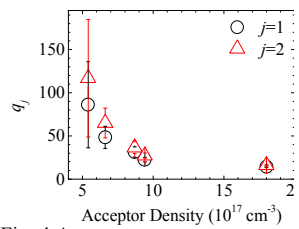

Fig. 4 Asymmetry parameter q_j obtained from the spectrum fitting

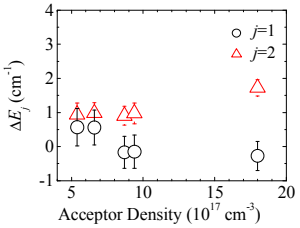

Fig. 5 The energy shift ΔE_j obtained from eq. (2).

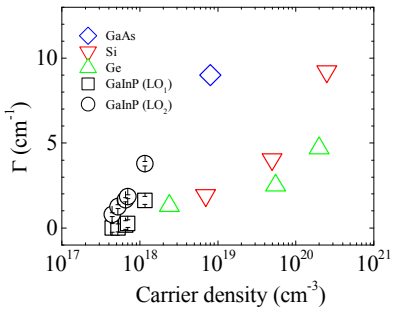

Fig. 6 Broadening parameters obtained from the spectrum fitting. For GaAs, 1 cm^{-1} [10] is subtracted as the original phonon broadening. In Si the original phonon broadening is included.

(a)), the broadening of LO_j phonon peak is not attributed to the degradation of the crystals. In Fig. 6, Γ values of other materials are also plotted [4-6]. It is known that Γ of polar semiconductors increases more rapidly than that of non-polar semiconductors as the hole density increases. The present result agrees with the general understanding that the electron-phonon interaction strength depends on the strength of the polarization of materials.

V. CONCLUSION

We have discussed quantum interference of two LO phonon modes and valence band states for p-$Ga_{0.5}In_{0.5}P$ films. As the hole density increases, asymmetric line shape, LO energy shift, and broadening were observed. The comparison of our result with that of one LO and one continuum system for other semiconductors reveals the difference in the strength of the polarization between polar and non-polar semiconductors. These results indicate that quantum interference of the two LO phonon modes and valence band states takes place in p-$Ga_{0.5}In_{0.5}P$ films. It is expected that this phenomenon would be a base of the control of absorption spectrum in the THz frequency region using the quantum interference of LO phonon-electron systems.

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