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# Raman scattering by interface-phonon polaritons in a GaAs/AlAs heterostructure

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We report the first Raman study of interface-phonon polaritons in an air/GaAs(60 nm)/AlAs(500 nm)/GaAs(substrate) system grown by molecular-beam epitaxy. Two interface-phonon polariton modes in the frequency region of the AlAs optical phonon, originating from the two (upper and lower) interfaces of GaAs/AlAs, were observed in a quasibackscattering configuration and in a nonresonance condition. The dispersion relations were obtained by changing the angle of the incident light. The experimental results agree well with the theoretical dispersion relation obtained from Maxwell's equations.

The surface and interface modes of elementary excitations are of considerable interest in solid-state physics.[1] Raman spectroscopy is a powerful tool to study such modes. Evans, Ushioda, and McMullen reported the first Raman study of double-interface surface-phonon polaritons (SPP's) in an air/GaAs(250 nm)/sapphire system.[2] The surface and interface-phonon modes lie between longitudinal optical (LO) and transverse optical (TO) phonon frequencies of active media, where the dielectric function $\epsilon(\omega)$ is negative. Recently, Raman scattering by interface-phonon modes in (Al,Ga)As superlattices has been investigated in resonance conditions, where the Fröhlich interaction enhances the Raman intensity of interface modes.[3-8] In those works, however, polariton properties were not discussed: Retardation effects, which produce polariton modes, were neglected in the calculation of dispersion relations and the wave-vector dependence of the mode frequency was not measured. Until now, there has been no report on experimental results of interface-phonon polariton (IPP) modes in (Al,Ga)As heterostructures.

In this Rapid Communication, we report the first Raman study of IPP modes in a GaAs/AlAs heterostructure grown by molecular-beam epitaxy (MBE): air/GaAs (thickness $d_1=60$ nm)/AlAs (thickness $d_2=500$ nm)/ GaAs substrate (semi-infinite) system. In this system, there are one SPP and two IPP modes of the GaAs optical phonon and two IPP modes of the AlAs optical phonon, where we define the SPP (IPP) mode as that originating from the air/GaAs(GaAs/AlAs) interface. The theoretical dispersion relations of such modes were calculated from Maxwell's equations following Ref. 9 by Mills and Maradudin. We have succeeded in measuring Raman scattering by the IPP modes in a nonresonance condition and in a quasibackscattering configuration, which was very weak, and obtained the experimental dispersion relation. Here, we discuss mainly the IPP modes of the AlAs optical phonon because the large frequency splitting between LO and TO phonon modes ($\sim 40$ cm$^{-1}$) enabled us to measure accurate frequencies.

The sample used in this work is a GaAs/AlAs heterostructure grown on a (001) semi-insulating GaAs substrate at 560 °C by MBE. The sample structure is GaAs($d_1=60$ nm)/AlAs($d_2=500$ nm)/GaAs (a 200-nm buffer layer and $\sim 300$-$\mu$m substrate). The Raman measurements were done in quasibackscattering configuration at room temperature. The incident light was a 514.5 nm line of an $Ar^+$ laser (nonresonance condition) with a typical power of 200 mW. The scattered light was analyzed by a Jobin-Yvon $U$-1000 double monochromator with a resolution of 1.5 cm$^{-1}$ and detected with a conventional photon counting system. The details of the scattering configuration, which gives the momentum of IPP mode, are described below.

Dispersion relations of SPP and IPP modes in an air($z>0$, $\epsilon_0=1$)/GaAs[$0>z>-d_1,\epsilon_1(\omega)$]/AlAs[$-d_1>z>-(d_1+d_2),\epsilon_2(\omega)$]/GaAs[$z<-(d_1+d_2),\epsilon_1(\omega)$] system can be obtained from Maxwell's equations.[9] The dielectric function $\epsilon_i(\omega)$ is given by

$$\epsilon(\omega)=\epsilon(\infty)+[\epsilon(0)-\epsilon(\infty)]\omega_T^2/(\omega_T^2-\omega^2)\,,$$

where $\omega_T$ is a TO phonon frequency. We set the $\alpha$ Cartesian component of the polariton electric field in the $x$-$z$ plane (TM wave) as

$$E_\alpha(x,z,t)=E_\alpha(z)\exp(iq_x x)\exp(-i\omega t)\,, \tag{1}$$

where the $x$ direction is parallel to the interfaces, and $q_x$ is the $x$ component of the polariton wave vector. We assume that the magnitude of $E_\alpha(z)$ falls to zero as $z\rightarrow\pm\infty$:

 

$$\begin{aligned}
&E_{0,x}(z)=A_0\exp(-q_{0,z}z) \quad \text{for } z>0(\text{air}),\\
&E_{1,x}(z)=A_1\exp(q_{1,z}z)+B_1\exp(-q_{1,z}z) \quad \text{for } 0>z>-d_1(\text{GaAs}),\\
&E_{2,x}(z)=A_2\exp(q_{2,z}z)+B_2\exp(-q_{2,z}z) \quad \text{for } -d_1>z>-(d_1+d_2)(\text{AlAs}),\\
&E_{3,x}(z)=A_3\exp(q_{1,z}z) \quad \text{for } z<-(d_1+d_2)(\text{GaAs}),
\end{aligned} \tag{2}$$

where $q_{i,z}$, which is given by $q_{i,z}=[q_x^2-\epsilon_i(\omega)(\omega^2/c^2)]^{1/2}$, is the $z$ component of the polariton wave vector in individual layers. The $z$ component of polariton electric field $[E_{i,z}(z)]$ is derived from $E_{i,x}(z)$ by using Maxwell's equations. From the boundary condition [continuity of $E_{i,x}(z)$ and $\epsilon_i(\omega)E_{i,z}(z)$ at $z=0$, $-d_1$, and $-(d_1+d_2)$], we get the following dispersion relation:

$$\left(1+\frac{q_1\epsilon_2}{q_2\epsilon_1}\right)\left[\left(1+\frac{q_0}{q_1}\epsilon_1\right)\left(1+\frac{q_2\epsilon_1}{q_1\epsilon_2}\right)-\exp(-2q_1d_1)\left(1-\frac{q_0}{q_1}\epsilon_1\right)\left(1-\frac{q_2\epsilon_1}{q_1\epsilon_2}\right)\right]$$
$$+\exp(-2q_2d_2)\left(1-\frac{q_1\epsilon_2}{q_2\epsilon_1}\right)\left[\left(1+\frac{q_0}{q_1}\epsilon_1\right)\left(1-\frac{q_2\epsilon_1}{q_1\epsilon_2}\right)-\exp(-2q_1d_1)\left(1-\frac{q_0}{q_1}\epsilon_1\right)\left(1+\frac{q_2\epsilon_1}{q_1\epsilon_2}\right)\right]=0, \tag{3}$$

where $q_i$ corresponds to $q_{i,z}$.

Figure 1 shows the theoretical dispersion relations (solid lines) of SPP and IPP modes in an air/GaAs(60 nm)/AlAs(500 nm)/GaAs system calculated from Eq. (3), where the dot-dashed lines indicate the photon dispersion relation: $\omega=ck_x/\epsilon_2(\omega)^{1/2}$ in the frequency region of the GaAs optical phonon and $\omega=ck_x/\epsilon_1(\omega)^{1/2}$ in that of AlAs optical phonon. The values of parameters used in this calculation are $\epsilon_1(0)=12.9$, $\epsilon_1(\infty)=10.9$, $\omega_{1,T}=269\,\text{cm}^{-1}$ for GaAs and $\epsilon_2(0)=10.1$, $\epsilon_2(\infty)=8.1$, and $\omega_{2,T}=361\,\text{cm}^{-1}$ for AlAs.[10] There are three polariton branches (G-1, G-2, and G-3) in the frequency region of the GaAs optical phonon and two polariton branches (A-1 and A-2) in that of the AlAs optical phonon. The arrows labeled $\omega_{I\text{-}1}$, $\omega_{I\text{-}2}$, and $\omega_{I\text{-}3}$ indicate the frequencies of those modes with $q_x\to\infty$: $\omega_{I\text{-}1}$ for $\epsilon_1=-\epsilon_2$, $\omega_{I\text{-}2}$ for $\epsilon_1=-1$, and $\omega_{I\text{-}3}$ for $\epsilon_2=-\epsilon_1$. Figure 2 shows the spatial distribution of $E_x(z)$ for the five polariton modes with the wave vector $q_x=8.5\times10^4\,\text{cm}^{-1}$ calculated from Maxwell's equations. From Fig. 2 we can obviously assign the origin of the polariton modes.

Figure 3 shows the Raman spectrum of the air/

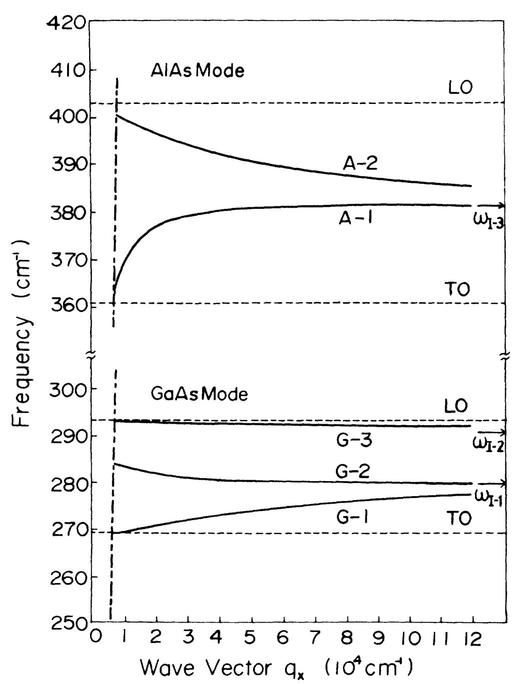


FIG. 1. Dispersion relations of SPP and IPP modes (solid lines) in an air/GaAs(60 nm)/AlAs(500 nm)/GaAs(substrate) system calculated from Eq. (3), where the dot-dashed lines are the photon dispersion relations. The arrows labeled $\omega_{I\text{-}1}$, $\omega_{I\text{-}2}$, and $\omega_{I-3}$ indicate the frequencies of the modes with $q_x\to\infty$.

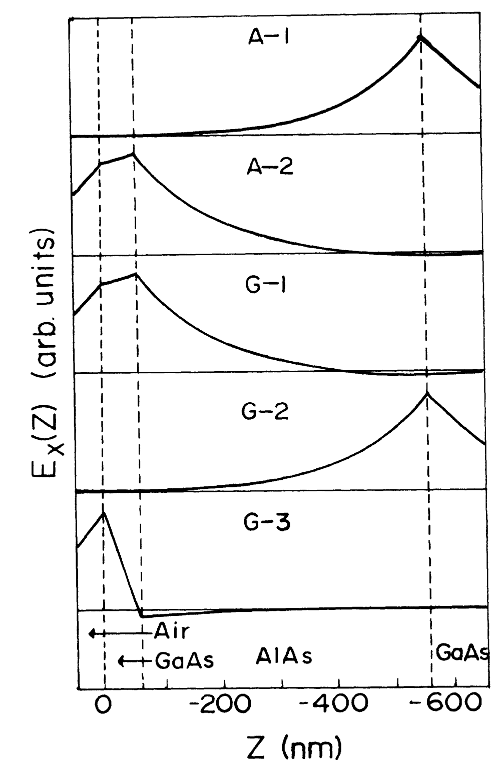


FIG. 2. Spatial distributions of $E_x(z)$ for the five polariton modes in the air/GaAs(60 nm)/AlAs(500 nm)/GaAs system with wave vector $q_x=8.5\times10^4\,\text{cm}^{-1}$ calculated from Maxwell's equations.

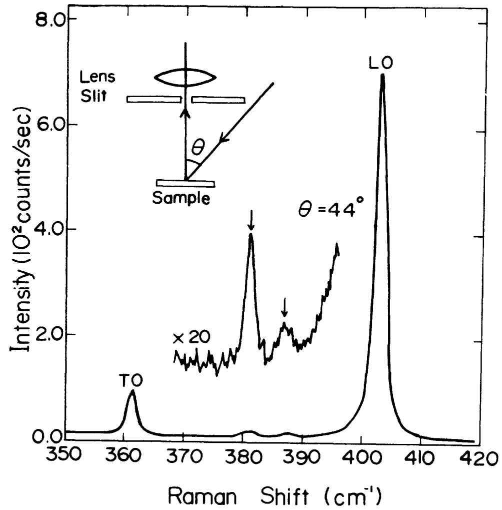


FIG. 3. Raman spectrum of the air/GaAs(60 nm)/AlAs(500 nm)/GaAs system in the frequency region of the AlAs optical phonon. The inset figure shows the measurement configuration (the incident-light angle $\theta = 44°$ in this case). The arrows indicate the frequencies of the IPP modes calculated from Eq. (3).

GaAs(60 nm)/AlAs(500 nm)/GaAs system in the frequency region of the AlAs optical phonon. The scattered-light polarization was not analyzed. The inset indicates the measurement configuration of Raman scattering. The incident-light angle $\theta$ (44° in this case) gives the value of wave vector $q_x = |\mathbf{k}_i| \sin\theta (8.48 \times 10^4\ \text{cm}^{-1})$, where $\mathbf{k}_i$ is the wave vector of incident light. A slit was set in front of the first input lens to reduce the uncertainty of the angle $\theta$: $\Delta\theta = \pm 1°$ in this experiment. In Fig. 3 the AlAs TO and LO phonons appear at 361 and 403 $\text{cm}^{-1}$, respectively. Moreover, two very small peaks are observed at 381 and 387 $\text{cm}^{-1}$. The arrows in Fig. 3 indicate the IPP frequencies of the AlAs optical phonon calculated from Eq. (3), which agree with the frequencies of the two small peaks; it is evident that the peaks correspond to Raman bands of the IPP modes. This is the first observation of IPP modes in the quasibackscattering configuration and in the nonresonance condition. The calculated spatial distribution of $E_x(z)$ shown in Fig. 2 indicates that the higher and lower frequency modes originate from the GaAs/AlAs interfaces at $z = -d_1$ and at $z = -(d_1 + d_2)$, respectively. The Raman scattering by the IPP modes was observed only in the case of $\mathbf{e}_i$ (polarization vector of incident light) perpendicular to $\mathbf{e}_s$ (that of scattered light); the selection rule is the same as that of the bulk phonon. This is consistent with the selection rule of SPP mode in an air/GaP system.[11]

For the IPP modes of the GaAs optical phonon, the Raman scattering was not clearly observed. The frequency splitting between the GaAs TO and LO phonons is about 20 $\text{cm}^{-1}$, which is half of the value of AlAs (about 40 $\text{cm}^{-1}$); it seems that the very weak Raman scattering by the IPP modes was hidden by the tail of LO phonon peak. It is expected that the IPP modes could be observed in a resonance condition, where the Raman intensity of the IPP modes should be remarkably enhanced due to the

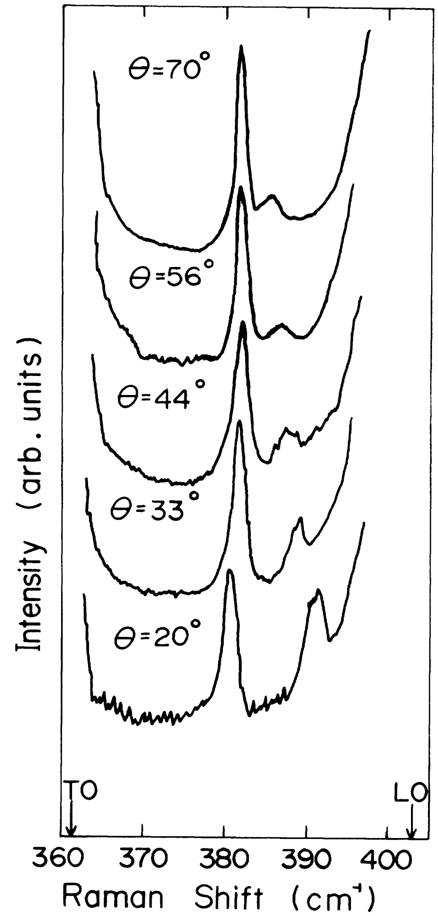


FIG. 4. Raman spectra of IPP modes of the AlAs optical phonon in the air/GaAs(60 nm)/AlAs(500 nm)/GaAs system with various incident-light angles.

Fröhlich interaction.[3-8]

Next we discuss the experimental results on dispersion relation of the IPP modes. The incident-light angle $\theta$ (Fig. 3) was varied from 7° to 70° to obtain the dispersion relation: the polariton wave vector ($q_x = |\mathbf{k}_i| \sin\theta$) from $1.49 \times 10^4$ to $1.15 \times 10^5\ \text{cm}^{-1}$. Figure 4 shows Raman

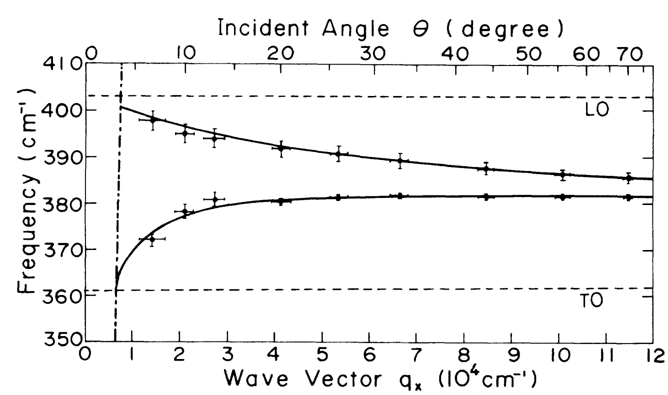


FIG. 5. Theoretical (solid lines) and experimental (closed circles) dispersion relations of IPP modes of the AlAs optical phonon in the air/GaAs(60 nm)/AlAs(500 nm)/GaAs system.

spectra of IPP modes in the frequency region of the AlAs optical phonon with various incident angles. The observed frequencies of the IPP modes depend on the angle $\theta$; this confirms that the two Raman bands correspond to the IPP modes. Figure 5 shows the theoretical and experimental dispersion relations of the IPP modes in the frequency region of the AlAs optical phonon: The solid lines indicate the theoretical results calculated from Eq. (3). It is evident that the experimental results fit in with the theoretical dispersion relations. Thus, the dispersion relation of IPP modes can be obtained in the quasibackscattering configuration.

In summary, we have performed the first Raman study of interface-phonon polaritons in an air/GaAs(60 nm)/AlAs(500 nm)/GaAs system. The theoretical dispersion relations derived from Maxwell's equations indicate that there are five polariton modes in this system: three (two) modes in the frequency region between the GaAs (AlAs) TO and LO phonon frequencies. In this work, two IPP modes of the AlAs optical phonon, which originate from the upper and lower interfaces of GaAs/AlAs, were observed in the quasibackscattering configuration and in the nonresonance condition. The experimental dispersion relations of the two IPP modes, which were obtained by changing an incident-light angle, agree well with the theoretical ones.

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