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Phonon replicas in the photoluminescence emission of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ alloys

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Phonon replicas in the photoluminescence spectra of a direct gap $\text{Al}_x\text{Ga}_{1-x}\text{As}$ alloy have been observed. The GaAs-like transverse optical and longitudinal optical as well as AlAs-like longitudinal optical modes were observed at the Γ -point. We also observe what we believe to be the longitudinal acoustical phonons at the L -point in the Brillouin zone. © 1995 American Institute of Physics.

In nonintentionally doped $\text{Al}_x\text{Ga}_{1-x}\text{As}$ with low Al concentration ($x < 0.125$) the free-exciton transition has been resolved.¹ When the x -value is increased, the free-exciton transition and bound exciton transitions are no longer resolved and the photoluminescence (PL) spectra of direct gap alloys consist of two main peaks. The higher energy peak is made up of unresolved free and bound exciton transitions; this peak is generally referred to as the bound exciton (BE) peak. The lower energy peak consists of conduction band to acceptor, free to bound (F-B) and donor to acceptor (B-B) transitions. These two characteristic emission peaks are shown in Fig. 1.

The phonon energies of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ have been obtained from lattice absorption measurements in the far-infrared, and from Raman spectroscopy.^{2,3} In the case of GaAs, the inelastic neutron scattering data were combined with various theoretical models to establish the lattice dynamics throughout the Brillouin zone. In regards to AlAs, few experimental data (only at zone center, and questionable at zone boundary) are presently available. To the best of our knowledge, an accurate theory capable of predicting lattice dynamics of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ throughout the Brillouin zone does not exist. Experimental measurements of the optical phonons in the ternary $\text{Al}_x\text{Ga}_{1-x}\text{As}$ alloy exhibit the two-mode behavior.² The two-mode behavior is typical of mixed crystals for which the optical bands of pure materials do not overlap and where, near the extremes of the composition range, the localized mode of the minority ions lies well outside the reststrahlen peak of the host crystal. By Kramers-Kronig transformation, the shift in frequency with Al concentration for the GaAs-like and AlAs-like longitudinal optical (LO) and transverse optical (TO) modes has been carefully studied. A two-mode behavior is observed for Raman peaks associated with LO phonons as well. Moreover, disorder-activated-transverse acoustic (DATA), longitudinal acoustic (DALA), and optical (DAO) bands are evidenced in resonant conditions, as caused by the relaxation of the crystal momentum conservation rule, due in turn, to the lack of periodicity. The energy dependence of these modes on the x -value of the alloy are usually determined from quadratic expressions.

We report here the observation of phonon replicas in the PL spectra of $\text{Al}_x\text{Ga}_{1-x}\text{As}$ which we interpret as phonons coupling to the free-exciton transitions. To our knowledge, this is the first report of phonon replicas of exciton transitions in direct gap $\text{Al}_x\text{Ga}_{1-x}\text{As}$ alloys to be observed in PL spectra. It is desirable to observe these transitions since they would provide an independent measure of the alloy composition. Previously, we have shown in GaAs that the phonons couple much more strongly to the free-exciton transitions than to the bound exciton transitions.⁴ We believe that the reason phonon replicas are not generally seen in the PL spectra of direct gap $\text{Al}_x\text{Ga}_{1-x}\text{As}$ alloys is that the bound exciton transitions are dominant over the free-exciton transitions. Assuming the alloy behaves similarly to GaAs the phonon coupling to the bound exciton transitions is considerably weaker than the coupling to the free-exciton transitions. We have coordinated the phonon energies observed in PL transitions to the calculated zone-center optical phonons obtained from the modified random element isodisplacement (MREI)

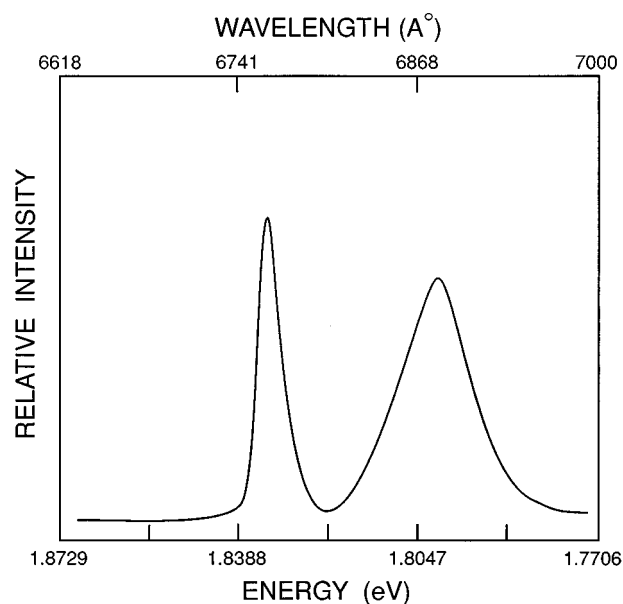


FIG. 1. Typical PL spectra for $\text{Al}_x\text{Ga}_{1-x}\text{As}$ direct gap material having an x value ≥ 0.2 .

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TABLE I. The physical parameters of binary compounds used in calculating the x -value dependence of optical phonons at 300 K in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ (with $A=\text{As}$, $B=\text{Ga}$, and $C=\text{Al}$). The calculations used the MREI model of Ref. 5.

Parameter	$\text{Al}_x\text{Ga}_{1-x}\text{As}$
$AB\omega_{\text{TO}}(\Gamma)$	268 cm^{-1}
$AB\omega_{\text{LO}}(\Gamma)$	292 cm^{-1}
ω_{loc}	362 cm^{-1}
$AB\varepsilon^\infty$	10.9
$AC\omega_{\text{TO}}(\Gamma)$	362 cm^{-1}
$AC\omega_{\text{LO}}(\Gamma)$	402 cm^{-1}
ω_{gap}	254 cm^{-1}
$AC\varepsilon^\infty$	8.5
F_{AB0}	$0.1773 \times 10^6\text{ dyn/cm}$
F_{AC0}	$0.1586 \times 10^6\text{ dyn/cm}$
F_{BC0}	$0.05 \times 10^6\text{ dyn/cm}$
Z_{AB}	0.514
Z_{AC}	0.624
θ	-0.15

model.⁵ The MREI model for the ternary alloy $\text{Al}_x\text{Ga}_{1-x}\text{As}$ can be summarized in terms of the following two fundamental assumptions: (i) like atoms vibrate with same amplitude and phase in the long-wavelength limit and (ii) the restoring forces acting on each atom are given by the statistical average of the interatomic forces between the neighboring atoms, assuming complete randomness. The interatomic force and the force due to the local electric field in a given alloy can be deduced from the macroscopic parameters characterizing the end binary members, such as dielectric constants and optical frequencies. The frequencies of the vibrational modes in $\text{Al}_x\text{Ga}_{1-x}\text{As}$ as a function of x are determined from the macroscopic parameters summarized in Table I. For $\text{Al}_x\text{Ga}_{1-x}\text{As}$, the model gives optical phonon energies at the zone center for the end member compounds (AlAs, GaAs) as well as their variation as a function of composition x . We observe the GaAs-like transverse optical (TO) and longitudinal optical (LO) as well as the AlAs-like longitudinal optical phonons. We also observe what we believe to be the longitudinal acoustical phonons at the L -point in the Brillouin zone. From the optical phonon replicas we can establish the free-exciton energy for the alloy.

The PL test structure consisting of a $1\text{-}\mu\text{m}$ -thick nominally undoped AlGaAs layer capped by 100 \AA of GaAs was grown by molecular beam epitaxy on semi-insulating (100) GaAs substrate. The AlGaAs layer was deposited at a rate of $0.6\text{ }\mu\text{m/h}$ at $\sim 680^\circ\text{C}$ using a V/III ratio of ~ 10 . Room-temperature Hall and conductivity measurements showed that the layer was p -type with a resistivity of $27\text{ }\Omega\text{ cm}$, hole mobility of $130\text{ cm}^2/\text{V s}$, and a hole concentration of $1.8 \times 10^{15}\text{ cm}^{-3}$. We observe sharp transitions on the low energy side of what is nominally referred to as the BE transition. These transitions are shown in Fig. 2, which contains the PL spectra from an $\text{Al}_x\text{Ga}_{1-x}\text{As}$ sample having a nominal x -value of 24%. What is usually observed in the literature³ are two unstructured peaks such as those shown in Fig. 1. We associate the structural features in Fig. 2 with phonon replicas of the BE peak. The BE peak is composed of

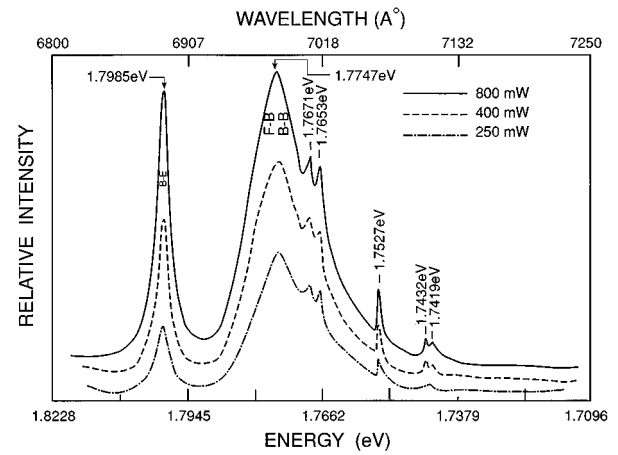


FIG. 2. Phonon replicas associated with the free exciton transition not resolved in the BE peak.

free-exciton transitions, donor-bound exciton transitions, and acceptor-bound exciton transitions. The peak at 1.7671 eV is associated with the GaAs-like TO phonon at the Γ -point, the peak at 1.7653 eV is associated with the GaAs-like LO phonon at the Γ -point, and the peak at 1.7527 eV is associated with the AlAs-like LO phonon at the Γ -point. The calculated energies of these three phonons for an x -value of 24% are 0.0337, 0.0352, and 0.0464 eV, respectively. Adding these energies to the energies of the respective phonon transitions, the following energies are obtained: 1.8008, 1.8005, and 1.7991 eV, the average of which is $1.8001 \pm 0.0009\text{ eV}$. We believe this energy to be that of the free exciton. This is consistent with our observation in GaAs which shows the phonon coupling to the free-exciton to be much stronger than the phonon coupling to the bound exciton. It is also noted that the linewidths of the phonon replicas are much narrower than the linewidth of the BE transition. This would follow if the phonons are primarily coupled to the free-exciton component of the BE transition. Two additional phonon replica peaks are observed at 1.7432 and 1.7419 eV. We associate these peaks with the LA phonon at the L -point, coupling to the GaAs-like TO and LO phonon peaks. In Ref. 4, a peak in the Raman spectra was observed near 0.0247 eV which was interpreted as the disorder-activated longitudinal acoustic mode. The peaks observed in Fig. 2 would place the energy of the LA mode at $0.0236 \pm 0.0003\text{ eV}$ in good agreement with the peak in the Raman spectra.³

In conclusion, we have observed phonon replicas in the PL spectra of an $\text{Al}_x\text{Ga}_{1-x}\text{As}$ alloy. We believe that the phonons are coupling to the free-exciton emission of the sample and that the phonon replica emissions are enhanced for this sample because the free-exciton emission is enhanced.

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