# Optical Phonons in $Ga_{1-x}Al_xAs$ Mixed Crystals: A Modified Random-Element Isodisplacement-Model Calculation

I. F. Chang
*Components Division, IBM Burlington, Essex Junction, Vermont* 05452
and
S. S. Mitra
*Department of Electrical Engineering, University of Rhode Island, Kingston, Rhode Island* 02881
(Received 27 March 1970)

The modified random-element isodisplacement model correctly predicts that $Ga_{1-x}Al_xAs$ forms a "two-mode"-type mixed crystal. The model calculations agree very well with a recent infrared measurement of concentration dependence of long-wavelength longitudinal and transverse optic mode frequencies of the mixed-crystal system.

The modified random-element-isodisplacement (MREI) model[1] has been successfully applied to many mixed-crystal systems. The model can predict, depending on the mass ratios, whether a given mixed crystal will exhibit "one-mode"- or "two-mode"- type behavior. For both types of mixed crystals the model can also correctly express the concentration dependence of the long-wavelength longitudinal optic (LO) and transverse optic (TO) mode frequencies. A number of examples from alkali halides and II–VI compounds have been furnished in Ref. 1. Calculations[1] have also been done on a III–V mixed crystal, viz., $GaP_{1-x}As_x$. However, comparison with experiment is incomplete because of limited experimental data. In this addenda, we present the MREI-model calculations on the mixed-crystal system $Ga_{1-x}Al_xAs$ to illustrate the applicability of this model to III–V compounds as well.

In some mixed crystals, e.g., those of zinc-blende–type crystals, the selection rules permit both $\vec{k} \sim 0$ LO and TO frequencies to be directly obtainable from Raman measurements.[2] In others, one can use the Kramers-Kronig dispersion analysis and/or the classical damped-oscillator fit of the reflection spectrum to obtain[3] the mode frequencies. Subsequently, it was pointed out[4] that instead of the so-called Drude's rule, a more rigorous method involving the damped-oscillator parameters may be used in obtaining the $\vec{k} \sim 0$ LO mode frequencies of the mixed crystals. For example, the LO and TO frequencies of $ZnS_{1-x}Se_x$[2,4] and $CdS_{1-x}Se_x$[3] as functions of composition were first obtained in this manner. In this type of presentation one can clearly see that the frequencies of the impurity modes, known as the local and gap modes in the case of two-mode-type mixed crystals emerge as triply degenerate limiting boundary values of LO and TO modes as $x \to 1$ or $x \to 0$. The splitting of the local or gap modes for small but nonzero values of $x$ or $1-x$ was also first experimentally demonstrated[2,3] in this manner.[5] A MREI-model calculation on $Ga_{1-x}Al_xAs$ will be presented in this form below.

Ilegems and Pearson[6] have recently measured the room-temperature infrared reflection spectra of $Ga_{1-x}Al_xAs$ for several values of $x$. They have obtained the TO and LO frequencies of the mixed-crystal system by the methods described above.[3] Two distinct reflection peaks with frequencies near those of pure AlAs and pure GaAs were observed for an intermediate value of $x$. Each band shifted to lower frequencies and decreased in intensity as the mole fraction of the corresponding component decreased. This clearly exhibits a two-mode-type behavior. The mass criterion derived from the MREI model, viz.,

$$m_{Al}(26.9) < \mu_{GaAs}(36.1), \tag{1}$$

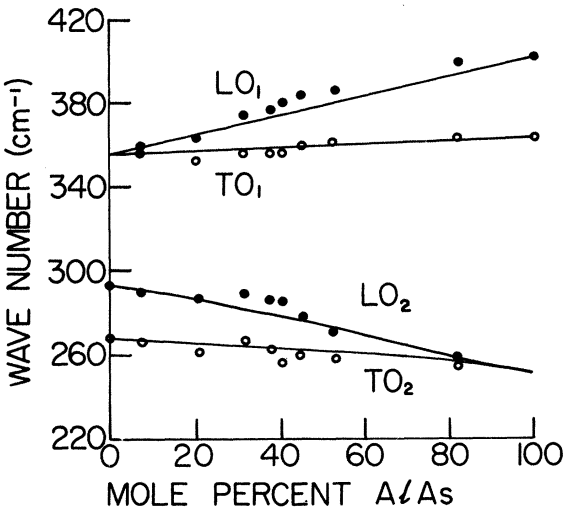


FIG. 1. Long-wavelength optic phonons in $Ga_{1-x}Al_xAs$. MREI-model calculations (solid lines) are compared with experimental data (closed and open circles) of Ref. 6.

TABLE I. MREI-model parameters for $Ga_{1-x}Al_xAs$.[a]

| | | | |
|---|---|---|---|
| Mass As, Al, Ga | | 74.92, | 26.98, 69.72 |
| $\epsilon_\infty$(AlAs), | $\epsilon_\infty$(GaAs) | 8.5 | 10.9 |
| $\omega_{AlAs}$(TO), | $\omega_{gap}$ | 364, | 252 |
| $\omega_{GaAs}$(TO), | $\omega_{loc}$ | 268, | 356 |
| $\omega_{AlAs}$(LO), | $\omega_{GaAs}$(LO) | 402, | 292 |
| $F_{AlAs0}$, | $F_{GaAs0}$, $F_{Al\text{-}Ga0}$, $\theta$ | 262.38, | 304.54 138.20 0.1484 |

[a] $\omega$ in $cm^{-1}$ and $F$ in $10^4$ $cm^{-2}$ $g$. For definition of symbols, see Ref. 1. Except for $\omega_{gap}$ and $\omega_{loc}$ frequencies, no other mixed-crystal data have been used in the calculation of parameters of last line.

where $m$ and $\mu$, respectively, denote atomic and reduced mass, indeed predict that $Ga_{1-x}Al_xAs$ is a two-mode system. Next, we perform a two-mode calculation based on the properties of the end member crystals. The derived parameters along with the physical properties of the crystals are given in Table I. Using the method of Ref. 1, the results obtained are presented in Fig. 1, where the experimental data of Ref. 6 are also shown. We conclude that (i) the MREI-model mass criterion predicts the correct mode behavior of the III–V mixed crystal $Ga_{1-x}Al_xAs$, and (ii) the model also predicts well the concentration dependence of the mode frequencies of this system.

We thank Professor G. L. Pearson for providing a preprint of Ref. 6.

[1]I. F. Chang and S. S. Mitra, Phys. Rev. 172, 924 (1968).

[2]O. Brafman, I. F. Chang, G. Lengyel, S. S. Mitra, and E. Carnall, Phys. Rev. Letters 19, 1120 (1967); in *Localized Excitations in Solids*, edited by R. F. Wallis (Plenum, New York, 1968), p. 602.

[3]J. Parrish, C. H. Perry, O. Brafman, I. F. Chang, and S. S. Mitra, in *II-VI Semiconducting Compounds*, edited by D. G. Thomas (Benjamin, New York, 1967), p. 1164.

[4]I. F. Chang, S. S. Mitra, J. N. Plendl, and L. C. Mansur, Phys. Status Solidi 28, 663 (1968).

[5]For a theoretical discussion of this splitting, see A. A. Maradudin and J. Oitmaa, Solid State Commun. 7, 1143 (1969).

[6]M. Ilegems and G. L. Pearson, Phys. Rev. B 1, 1576 (1970).

PHYSICAL REVIEW B VOLUME 2, NUMBER 4 15 AUGUST 1970

# ERRATA

**Relativistic Band Structure and Electronic Properties of SnTe, GeTe, and PbTe**, Y. W. Tung and M. L. Cohen [Phys. Rev. 180, 823 (1969)]. The estimate for the spin-orbit splitting for PbTe at $\Gamma$ [p. 824, fourth line after Eq. (2)] should be 1.09 eV and not 1.82 eV. This smaller splitting results in an upward shift of the third valence band in Fig. 3; the shape of the band is unchanged. The shifts of the energies for this band is the symmetry points $\Gamma$, $X$, $L$, $K$ are 0.66, 0.45, 0.73, and 0.38 eV, respectively.

**Optical Phonons in Sodium Chlorate**, C. M. Hartwig, D. L. Rousseau, and S. P. S. Porto [Phys. Rev. 188, 1328 (1969)]. In Table I, the 65 $cm^{-1}$ line in the $A$ symmetry column should be in the $E$ symmetry column. The 86 $cm^{-1}$ line in the $E$ symmetry column should be in the $A$ column.