Localization of a disordered phonon system: Anderson localization of optical phonons in $Al_xGa_{1-x}As$

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Localization properties of an isotopically disordered phonon system are studied by mapping the system to an electronic tight-binding problem with on-site disorder. When the model parameters are adjusted for the $Al_xGa_{1-x}As$ alloys, it is shown that there exists x_c such that for $x > x_c$, all GaAs-like optical-phonon states are localized while for $x < x_c$, most states are extended except for some near the edges of the GaAs-like branch. These results are confirmed by an independent numerical investigation of the scaling behavior of the localization lengths. Our results reconcile current controversy regarding the nature of GaAs-like optical-phonon eigenstates in the $Al_xGa_{1-x}As$ alloys. [S0163-1829(96)51628-0]

Ever since the famous paper of Anderson,[1] the localization transition in disordered systems has been one of the main areas of research. For phonon systems, however, relatively few results are available while a great amount of work exists for electronic systems. So far, a clear quantitative connection between these two systems is not yet established.

As a related problem in a real situation, the optical-phonon property in $Al_xGa_{1-x}As$ alloys is an important issue where some controversies exist. The asymmetric Raman profiles of the GaAs-like (GL) branch have been fitted on the assumption of finite [$\lesssim 100$ Å] phonon mode correlations[2] while a recent paper reports that well-defined GL optical-phonon dispersions exist for all values of x.[3] These two results are somewhat inconsistent with each other since a well-defined dispersion relation itself may imply that the spatial correlation function of the phonon is infinite in extent. Needless to say, the word ''dispersive'' should be taken with caution, since the inherent uncertainty in **k** (Δk) due to alloy fluctuation results in line broadening and limits the spatial extent to $\sim 1/\Delta k$.[4,5]

In this paper, a simple model for phonons of a ternary alloy system is constructed and it is shown that, by an appropriate identification of corresponding parameters, our phonon model is exactly mapped to an Anderson model generalized to a diatomic basis lattice. This enables us to study the localization property of the isotopically disordered phonon system by the same method as that used for the Anderson model; in the present calculation we use the method introduced by Licciardello and Economou.[6]

As for the $Al_xGa_{1-x}As$ alloys, the results obtained within our model calculation present a general localization picture that reconciles two previously mentioned, seemingly contradictory results. It is shown that above $x_c \cong 0.45$, all GL optical-phonon states are localized, which is in agreement with the assumption of Parayanthal and Pollak.[2] For $x < x_c$, most states are extended except for some near the edges of the GL branch, in which case a dispersion relation may still be a useful description of the eigenstates. Thus in our picture

the previous two seemingly conflicting results remain valid as two limiting cases depending on whether $x < x_c$ or $x > x_c$. Our main results are confirmed by an independent numerical study of the scaling behavior of the localization lengths.

We consider an alloy of chemical formula $A_{1-x}B_xC$ on the zinc-blende structure. A cation site of the lattice is randomly occupied by an A or a B atom with probability $(1-x)$ or x, respectively. For simplicity, it is assumed that a displacement from its equilibrium position can be represented by a scalar quantity and that the elastic interactions are restricted only to those with nearest-neighbor (nn) sites. The equations of motion for this system are written as follows:

$$M_i \frac{d^2 U_i}{dt^2} = K \sum_j^i (u_j - U_i),$$

$$m_i \frac{d^2 u_i}{dt^2} = K \sum_j^i (U_j - u_i), \quad (1)$$

where M_i (m_i) and U_i (u_i) are the mass and the atomic displacement from equilibrium position of the atom at the ith cation (anion) site, respectively. The superscript i of the sum denotes that the summation is over only the nn sites of the site i. There is no mass randomness on the anion sites so that we can set m_i's all equal to M_C. The relations $U_i = \widetilde{U}_i e^{-i\omega t}$ and $u_i = \widetilde{u}_i e^{-i\omega t}$ are inserted into Eq. (1) and each equation is divided by ω^2. Then we get the equations of following form:

$$(\epsilon_i - E)\widetilde{U}_i + V(E)\sum_j^i \widetilde{u}_j = 0,$$

$$(\epsilon - E)\widetilde{u}_i + V(E)\sum_j^i \widetilde{U}_j = 0, \quad (2)$$

where we have changed the notations of parameters; $M_i \to \epsilon_i, M_C \to \epsilon, zK/\omega^2 \to E$ and $K/\omega^2 \to V$. $z = 4$ is the coordination number of the diamond lattice. We find that Eq. (2) is the eigenvalue equations of the Anderson model generalized to a diatomic basis lattice. The quantities corresponding to the on-site energies of one sublattice are randomly distributed according to a binary distribution of the mass of atom A (M_A) and the mass of atom B (M_B). On the other hand, it should be noted that since the hopping energy V is dependent on E, for two distinct values of E, two corresponding Anderson Hamiltonians have hopping energies different from each other.

Now we adopt the localization criterion introduced by Licciardello and Economou.[6] The single-particle Green's function shows different analytic behavior depending on whether the eigenstate of energy E is an extended state or a localized one; the imaginary part of the self-energy goes to zero for a localized state while it remains nonzero for the extended state when E approaches the real axis in the complex E plane.[7] When the self-energy is expanded into a perturbation series with respect to the hopping energy, the localization function $L(E)$ is obtained as the ensemble-averaged value of the limiting term of the series. The numerical value of $L(E)$ determines the nature of the eigenstate of energy E; for $L(E) \gtrsim 1$ (<1), the series for the self-energy diverges (converges) and the corresponding state is an extended (a localized) one. A generalization of their result, which is for a monatomic system, to our diatomic one gives the localization function as follows:

$$ L(E) = \lim_{M \to \infty} \left[V^{M+1} \sum \, (\widetilde{G}_\alpha)_1^0 (\widetilde{G}_c)_2^{0,1} \cdots (\widetilde{G}_a)_M^{0,1,\ldots,M-1} \right]^{1/M} . \tag{3} $$

The summation is over all indices $1, 2, \ldots, M$ with the restrictions corresponding to all self-avoiding paths of order M starting and ending at the site 0. The quantity $(\widetilde{G}_{a(c)})_i^{0,1,\ldots,i-1}$ is defined by

$$ \ln(\widetilde{G}_{a(c)})_i^{0,1,\ldots,i-1} \equiv \langle \ln |(G_{a(c)})_i^{0,1,\ldots,i-1}| \rangle. \tag{4} $$

$(G_{a(c)})_i^{0,1,\ldots,i-1}$ is the anion (cation) diagonal element of the Green's function at site i for Eq. (2) with the sites $0, 1, \ldots, i-1$ excluded. The $L(E)$ given by Eq. (3) is very complicated for practical calculations. On the other hand, an estimate $L^*(E)$ is found by the following approximations:[6]

$$ L(E) \cong VK |(\widetilde{G}_a)_1^0 (\widetilde{G}_c)_2^1|^{1/2} \cong VK |(\overline{G}_a)_1^0 (\overline{G}_c)_2^1|^{1/2} \equiv L^*(E), \tag{5} $$

where K is the connectivity,[8] which is evaluated to be $\cong 2.88$ for the diamond lattice. The second approximation of Eq. (5) is replacing the Green's functions defined in Eq. (4) by those obtained by the coherent potential approximation (CPA).[6]

For ordered cases, i.e., $x = 0$ or $x = 1$, $L(\omega)$ is equal to 1 at the edges and larger than (smaller than) 1 inside (outside) the branches. However, due to the approximations involved in Eq. (5), in general, our estimate $L^*(\omega)$ does not satisfy these conditions. Therefore a weighting function $W(\omega, x)$ is introduced so that the product of $W(\omega, x)$ and
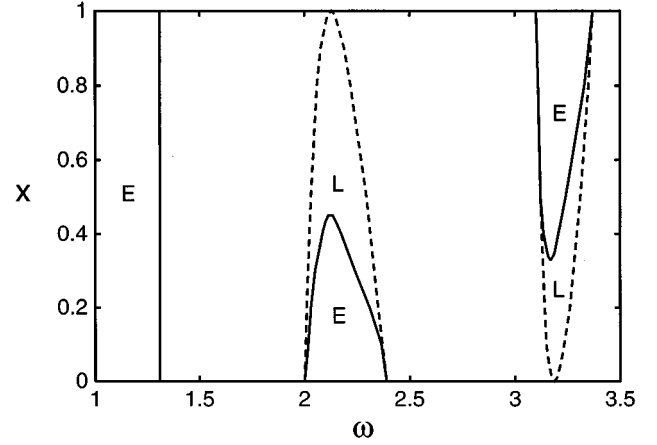

FIG. 1. The phase diagram drawn in the ω-x plane for $\text{Al}_x\text{Ga}_{1-x}\text{As}$. ''$L$'' represents the region of localized eigenstates and ''E'' the region of extended eigenstates, the solid lines forming the boundaries between these two regions. The dashed lines represent the trajectories of phonon branches within the CPA. For the acoustic branches the dashed line nearly coincides with the solid line.

$L^*(\omega)$ would be a correct estimate of $L(\omega)$ at the edges of the branches.[9] We may use,[9] as a weighting function,

$$ W(\omega, x) = \left(A \exp\left[-\frac{(\omega^2 - z(K/M_C + \eta)/2)^2}{\Delta^2} \right] + 1 \right)^{-1}, \tag{6} $$

where A and Δ are fitting parameters to satisfy condition (iii) of Ref. 9 and $\eta \equiv (1-x)(K/M_A) + x(K/M_B)$.

The results of calculation for $K/M_B = 2.41$ and $K/M_C = 0.43$ with K/M_A normalized to 1 are shown in Fig. 1; these numerical values have been obtained by fitting the optical-phonon spectral ranges of two pure materials of our model, i.e., AC and BC, to the reported experimental data of pure GaAs and pure AlAs, respectively.[10,11] In what follows, numerical values of ω normalized by $\sqrt{K/M_A}$ are used. We interpret the states in the frequency range between 2.0 and 2.39 (238 and 285 in cm^{-1}, respectively) as the GaAs-like optical-phonon branch and the ones between 3.1 and 3.37 (370 and 404 in cm^{-1}, respectively) as the AlAs-like optical-phonon branch. As x increases, the states of the GL optical branch localize because the Al atoms are ''barriers'' for these excitations while those of the AlAs-like optical branch localize as $(1-x)$ increases. Restricted to the GL branch, all phonon excitations localize beyond $x_c \cong 0.45$; as x increases, localization proceeds from the edges and the boundaries between the extended states and the localized states move inward until they coalesce at x_c.

Variation of x_c over some range of the parameters K/M_B and K/M_C is shown in Fig. 2. Within such a range of input parameters, qualitative behaviors of optical-phonon branches are not different from those of Fig. 1, although the numerical value of x_c varies. x_c is a monotonically decreasing (increasing) function of K/M_B (K/M_C). One can explain such a behavior intuitively when the variation of the gap size between the two optical-phonon branches of the pure materials is considered; the gap size is given as $\sqrt{zK/M_B} - \sqrt{z(1+K/M_C)}$, which is monotonically increasing (decreasing) with respect to K/M_B (K/M_C). As the gap size increases, the probability that an GL optical-phonon ex-
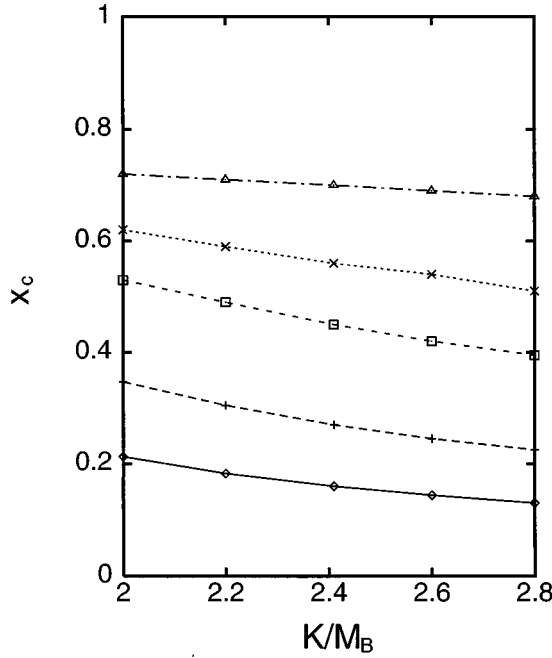
FIG. 2. Variation of x_c as a function of K/M_B and K/M_C. $K/M_C=0.2$ ($\diamond$), 0.3 (+), 0.43 ($\square$), 0.5 ($\times$), and 0.6 ($\triangle$) from bottom to top.

citation can penetrate the site occupied by an Al ion gets smaller since there is a larger difference between the natural vibrational frequencies of two species. Therefore GL optical-phonon excitation localizes more easily, i.e., yields smaller x_c, at a larger gap size.

It should be noted that in Eq. (5) we approximate the geometric average by the CPA one. Though such procedure has been shown to yield qualitatively correct results for localization transition in the three-dimensional (3D) Anderson model,[12] its validity for other general 3D systems is not established yet. Therefore, we perform an independent calculation to confirm our results for the ternary alloy system.

The same form of equations of motion as Eq. (1) is considered but on the NaCl structure instead of the zinc-blende structure, for simplicity. The localization length along the z direction, λ_M, for a bar-shaped geometry of $M \times M \times N$ is calculated from the following formula:

$$\frac{1}{\lambda_M(\omega,x)} = -\lim_{N\to\infty}\frac{1}{N}\ln|G(1,N;\omega,x,M)|, \quad (7)$$

where $G(1,N;\omega,x,M)$ designates the Green's function coupling pairs of masses at opposite ends of the bar of length N. We calculate the Green's function recursively by the method of MacKinnon and Kramer[13] for $M \leq 14$. A periodic boundary condition has been imposed in the x and y directions and only even numbers of M have been considered in our calculation.

In Fig. 3(a), the results for the logarithm of the renormalized localization length, $\Lambda(\omega,x) \equiv \lambda_M/M$ for $\omega = \sqrt{7} \cong 2.65$ are presented as a function of x for several values of M. Note that since the coordination number of the NaCl structure is 6, for the same input parameter values, i.e., $K/M_B=2.41$ and $K/M_C=0.43$, the GaAs-like branch ranges
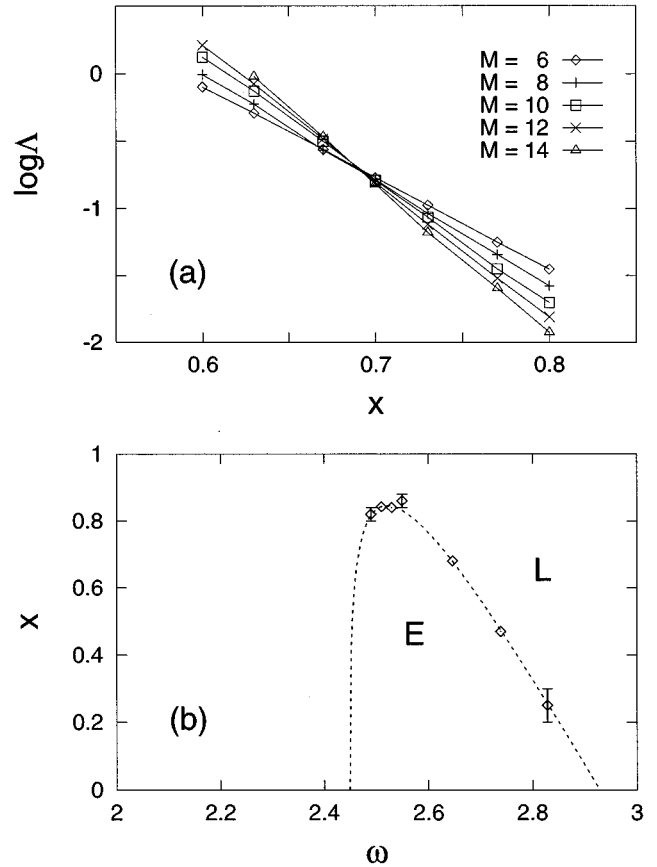

FIG. 3. (a) Typical behavior of the renormalized localization length Λ as a function of x for M ranging from 6 to 14. The intersection point, which is interpreted as the transition point, is estimated as 0.68 ± 0.01 for this data set. (b) The phase diagram drawn in the ω-x plane. The squares represent the data points for the critical concentrations and the dashed line is a fitting curve to the data points. "L" represents the region of localized states and "E" the region of extended states.

from $\omega = \sqrt{6} \cong 2.45$ to $\omega \cong 2.93$. With rising M, Λ increases for smaller values of x while for larger values of x it keeps decreasing. This implies that in the macroscopic limit, i.e., $M \to \infty$, the vibrational excitation at $\omega \cong 2.65$ goes from an extended state to a localized state as x increases, e.g., from 0.6 to 0.8. The localization transition is identified as the intersection point of the curves in Fig. 3(a). For the data set presented in Fig. 3(a), the transition concentration is estimated to be 0.68 ± 0.01.

Similar procedures for some other values of ω within the GL branch, i.e., from $\omega \cong 2.45$ to $\omega \cong 2.93$, have been performed and the resulting phase diagram is depicted in Fig. 3(b). The squares represent the data points for the critical concentrations and the dashed line is a fitting curve to the data points with the restriction of being zero at the edge of the branch. Figure 3(b) shows that the qualitative picture we have obtained by the previous analytic calculations is correct though the numerical value of $x_c=0.85 \pm 0.01$, i.e., the concentration beyond which the whole excitation of the GL branch localizes, is somewhat larger. This result is natural since the coordination number of the NaCL structure, i.e., 6, is larger than that of the zinc-blende structure, i.e., 4, and the system with more nn's will prefer the extended state. Assum-

ing simple linear dependence with respect to the coordination number, we expect that the calculation on the zinc-blende structure may yield $x_c \cong 0.85 \times (4/6) \cong 0.57$, close to $x_c \cong 0.45$ from the previous analytic results.

Therefore, from both the analytic and numerical methods, we contend that there exists some nontrivial x_c such that when $x > x_c$ all GL optical phonons are localized, justifying the method of Parayanthal and Pollak.[2] On the other hand, when $x < x_c$, most GL optical phonons are extended while the states near the edges of the branch are localized. In the latter case, a dispersion relation $\omega = \omega(\mathbf{k})$ may be a useful description of the eigenstates. However, the phrase ''dispersion relation'' here should be interpreted in a rather different sense from an ordered system; an eigenstate of energy ω is a superposition of *many different* Bloch states whose distributions $P_\omega(\mathbf{k})$ are centered at $\mathbf{k}$ but which have some finite linewidth caused by incoherent scattering from the disordered assembly of sites. This linewidth is given as $\sim 1/\xi$ if the eigenstate is localized within a spatial extent ξ. Therefore in a disordered system the existence of a dispersion relation does not necessarily imply that all eigenstates are extended. One can safely use the terminology of dispersion relation in a sense qualitatively the same as an ordered system only when a significant fraction of eigenstates of given branch are extended.

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[9] To determine $W(\omega,x)$ the following properties of $L^*(\omega)$ should be noted: First, in the limit of $E \to \infty$, both $L^*(E)$ of Eq. (5) and $L(E)$ of Eq. (3) approach VK/E. This implies that the weighting function should approach unity as ω increases to infinity. Second, for ordered cases, all the Green's functions of Eq. (3) and Eq. (5) are parametrized by $(\omega^2 - zK/M_C)$ $(\omega^2 - zK/M_{A(B)})/(zK^2/M_C M_{A(B)})$, so that both $L(\omega)$ and $L^*(\omega)$ turn out to be symmetric functions of ω^2 with respect to $\omega^2 = zK(1/M_C + 1/M_{A(B)})/2 \equiv \omega^2_{A(B)}$ for $x = 0$ $(x = 1)$. Third, $L^*(\omega)$ yields constant numerical values at the edges of the branches; $L^*(\omega) \cong 1.04$ at $\omega^2 = 0$ and $\cong 1.33$ at $\omega^2 = zK/M_{A(B)}$, independent of the input values of M_A, M_B, and M_C. Therefore the weighting function should satisfy the following conditions: (i) $W(\omega,x) \to 1$ as $\omega \to \infty$, (ii) for $x = 0$ or $x = 1$, $W(\omega,x)$ is even with respect to $\omega_{A(B)}$ as a function of ω^2, and (iii) $W(0,x) = 1/1.04$ and $W(\sqrt{zK/M_{A(B)}},x) = 1/1.33$. Therefore on the assumption that the evenness of the weighting function is also true for the disordered cases and the center of symmetry varies linearly with x, we may use the weighting function shown in the text.

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