Renormalization-Group Method for Vibrational Behavior in Mixed Diatomic Crystals

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The renormalization-group method has been applied to investigate vibrational properties of a diatomic mixed crystal. It has been found that there exists a fixed point which separates the one-mode behavior from the two-mode behavior. This transition depends on concentration, force constants, and mass ratios.

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It has been shown that a fixed point exists which separates the extended mode behavior from a localized one which is interpreted as a transition from the one-mode to two-mode behavior.[1] Several models have been developed in order to find a criterion which separates the one-mode behavior from the two-mode one.[1] Among these the coherent-potential approximation (CPA),[2] the modified random-element-isodisplacement model, (MREI),[3] the $n \to 0$ trick,[4] and the recursion method[5] have been applied. We propose here a new criterion based on the renormalization-group[6] (RG) method, which is in agreement with experimental[1] work and also with existing criteria[2,3] (Fig. 1).

Our RG transformation consists in comparing the eigenvalues, coupling constants, and probability distribution of the initial lattice with those of a new one of spacing S ($S>1$) times larger than the original. We choose to describe the given chain of atoms with randomly distributed masses m_B and m_C first as a chain of cells with two atoms, and afterwards as a chain of cells of four atoms. We are interested in the long-wave optical mode. Therefore, after computing the eigenvalue and the eigenvector for each basic cell, we preserve only the long-wave optical phonons, drop out the acoustical mode, and obtain our initial Langrangian with $S=1$ which is given in Eq. (2d). Then we will write the coupling between atoms in different cells as a coupling between cells.

The Lagrangian of the chain of two-atom cells is given by

$$\mathcal{L}_0 = \tfrac{1}{2}[(\epsilon_1 r - 2)x_1^2 + (r-2)x_2^2 + 4x_1x_2] + x_2(x_3 - x_1) + \tfrac{1}{2}[\cdots, \tag{1}$$

where

$$r \equiv \omega^2/(K_x^x/m_A), \quad \epsilon_l \equiv m_2(l)/m_A, \quad l = 1, 2, 3,$$

ω is the frequency, m_A is the constant mass, and $m_2(l)$ is m_B with probability z and m_C with probability $1-z$. [ϵ_l takes two values; $m_B/m_A = \epsilon$, $m_C/m_A = \epsilon(1-\delta)$ with $0<\delta<1$.]

We have two mass configurations, $\sigma_1 = \{m_B, m_A\}$ and $\sigma_2 = \{m_C, m_A\}$, and, respectively, two eigenvalues, r_1 and r_2, and two eigenvectors, $\Psi\sigma_1$ and $\Psi\sigma_2$:

$$r_1 = 2(l+\epsilon)/\epsilon, \quad \Psi\sigma_1 = (1+\epsilon^2)^{-1/2}(x_1 - \epsilon x_2); \tag{2a}$$

$$r_2 = 2[1+\epsilon(1-\delta)]/\epsilon(1-\delta), \quad \Psi\sigma_2 = [1+\epsilon^2(1-\delta)^2]^{-1/2}[x_1 - \epsilon(1-\delta)x_2]. \tag{2b}$$

The probability distribution of the eigenvalues is

$$P(r) = Z\delta_{r_1 r_2} + (1-Z)\delta_{r_1 r_2}. \tag{2c}$$

The nondiagonal term written using Eqs. (2a), (2b), and (1) becomes

$$\mathcal{L} = \sum_{\alpha=1}^{N/2} \{\tfrac{1}{2}[r - v_\sigma(\alpha)]\Psi^2(\alpha) + t_{\sigma,\sigma'}{}^{\alpha,\alpha+1}\Psi(\alpha)\Psi(\alpha+1)\}. \tag{2d}$$

α is the cell index, $v_\sigma(\alpha)$ is the diagonal term depending on the configuration, and $t_{\sigma,\sigma'}{}^{\alpha,\alpha+1}$ is the coupling term between the cells α and $\alpha+1$ which depends on the configuration σ_1, σ_2.

The Lagrangian given in Eq. (1) is then written in a form of four atoms per cell and is given by

$$\mathcal{L}_0 = \tfrac{1}{2}[(\epsilon_1 r - 2)x_1^2 + (r-2)x_2^2 + (\epsilon_3 r - 2)x_3^2 + (r-2)x_4^2 + 2x_1x_2 + 2x_2x_3 + 2x_3x_4 + 2x_1x_4]$$
$$+ x_4(x_5 - x_1) + \tfrac{1}{2}[\cdots. \tag{3}$$

We have now four configurations:

$$\sigma_1 \equiv \{m_1 = m_3 = m_B\};\quad \sigma_2 \equiv \{m_1 = m_3 = m_C\};\quad \sigma_3 \equiv \{m_1 = m_B;\ m_3 = m_C\};\quad \sigma_4 \equiv \{m_1 = m_C;\ m_3 = m_B\}.$$

In order to perform our RG transformation we are going to keep, in the four-atom cell, only the mode corresponding to the configuration of two optical modes in the two-atom cells. This choice corresponds to calculating the optical mode from the two optical modes in the two-atom cells. The spacing of the new problem is $S = 2$. We obtain the following configurations for the chosen optical mode:

$$r_1 = 2(1+\epsilon)/\epsilon,\quad \Psi\sigma_1 = [2(1+\epsilon^2)]^{-1/2}(x_1 - \epsilon x_2 + x_3 - \epsilon x_4); \tag{4a}$$

$$r_2 = \frac{2[1+\epsilon(1-\delta)]}{\epsilon(1-\delta)},\quad \Psi\sigma_2 = \frac{x_1 - \epsilon(1-\delta)x_2 + x_3 - \epsilon(1-\delta)x_4}{[1+\epsilon^2(1-\delta)^2]^{1/2}}; \tag{4b}$$

$$r_3 = \left(\frac{2-\delta}{\epsilon(1-\delta)} + 1\right) + \left[\left(\frac{2-\delta}{\epsilon(1-\delta)} + 1\right)^2 - 4\left(\frac{\epsilon + 1 - \frac{1}{2}\epsilon\delta}{\epsilon^2(1-\delta)}\right)\right]^{1/2}, \tag{4c}$$

$$\Psi\sigma_3 = (1 + a^2 + 2b^2)^{-1/2}(ax_1 + bx_2 + x_3 + bx_4),\qquad a \equiv 1 + \epsilon\delta r_3/(2 - r_3\epsilon),\quad b \equiv 1 - \tfrac{1}{2}\epsilon r_3(1-\delta);$$

$$r_3 = r_4,\quad \Psi\sigma_4 = (1 + a^2 + 2b^2)^{-1/4}(x_1 + bx_2 + ax_3 + bx_4). \tag{4d}$$

The probability distribution in this case is given by

$$P'(r') = Z^2\delta_{r',r_1} + (1-z)^2\delta_{r',r_2} + Z(1-Z)\delta_{r',r_3} + Z(1-Z)\delta_{r',r_4}. \tag{4e}$$

As in the case with two atoms we obtain

$$\mathfrak{L}' = \sum_{\alpha=1}^{N/2}\left[\tfrac{1}{2}(r' - v_\sigma'(\alpha)\Psi'^2(\alpha) + (t')_{\sigma,\sigma'}{}^{\alpha,\alpha+1}\Psi'(\alpha)\Psi'(\alpha)\right]. \tag{4f}$$

We replace[7] $t_{\sigma,\sigma'}{}^{\alpha,\alpha+1}$ and $(t')_{\sigma,\sigma'}{}^{\alpha,\alpha+1}$ by $t_{\rm eff}{}^{\alpha,\alpha+1} = \langle | t_{\sigma,\sigma'}{}^{\alpha,\alpha+1} | \rangle_P$ and $(t')_{\rm eff}{}^{\alpha,\alpha+1} = \langle | (t')_{\sigma,\sigma'}{}^{\alpha,\alpha+1} \times | \rangle_{P'}$. By replacing $t_{\sigma,\sigma'}{}^{\alpha,\alpha+1}$ by $t_{\rm eff}$, the randomness in the Lagrangian is reduced so that the "true" randomness is probably larger than what our estimate will yield. Computing $t_{\rm eff}$ and $t_{\rm eff}'$ we obtain

$$t_{\rm eff} = \frac{\epsilon(2-\delta)Z(1-Z)}{[1+\epsilon^2(1-\delta^2)]^{1/2}(1+\epsilon^2)} + \frac{\epsilon}{1+\epsilon^2}Z^2 + \frac{\epsilon(1-\delta)Z(1-Z)}{1+\epsilon^2(1-\delta)^2}, \tag{5a}$$

$$\begin{aligned} t_{\rm eff}' = \Bigg[&\left(\frac{2|ab|+2|b|}{1+a^2+2b^2} + \frac{\epsilon(1-\delta)+\epsilon}{2(1+\epsilon^2)^{1/2}[1+\epsilon^2(1-\delta)^2]^{1/2}}\right)Z^2(1-Z)^2 + \frac{\epsilon}{(1+\epsilon^2)}Z^4 \\ &+ \frac{\epsilon(1-\delta)}{2[1+\epsilon^2(1-\delta)^2]}(1-Z)^4 + \frac{2|b|+\epsilon(|a|+1)}{(1+a^2+2b^2)^{1/2}[2(1+\epsilon^2)]^{1/2}}Z^3(1-Z) \\ &+ \frac{2|b|+\epsilon(1-\delta)(|a|+1)}{(1+a^2+2b^2)[2(1+\epsilon^2)(1-\delta)^2]^{1/2}}Z(1-Z)^3\Bigg]. \end{aligned} \tag{5b}$$

We define $E_\sigma(\alpha)$ and $E_\sigma'(\alpha)$:

$$E_\sigma(\alpha) \equiv [r - v_\sigma(\alpha)]/t_{\rm eff};\quad E_\sigma'(\alpha) \equiv [r - v_\sigma'(\alpha)]/t_{\rm eff}'. \tag{6}$$

The Lagrangians $\mathfrak{L}$ and $\mathfrak{L}'$ become

$$\mathfrak{L} = \sum_{\alpha=1}^{N/2}\left[\tfrac{1}{2}E_\sigma(\alpha)\Psi^2(\alpha) + \Psi(\alpha)\Psi(\alpha+1)\right], \tag{7}$$

$$\mathfrak{L}' = \sum_{\alpha=1}^{N/4}\left[\tfrac{1}{2}E_\sigma'(\alpha)\Psi'^2(\alpha) + \Psi'(\alpha)\Psi'(\alpha+1)\right]. \tag{8}$$

It has been shown[4,8] that the density of states is related to $\langle \ln Z\rangle_P$ (where Z is expressed as a functional integral of the Lagrangian). Assuming that the density of states is invariant, we have

$$\lim_{n\to 0}\left(\frac{\langle Z^n\rangle_P - 1}{n}\right) = \left(\frac{\frac{1}{2}N}{\frac{1}{4}N}\right)\lim_{n\to\infty}\left(\frac{\langle Z^n\rangle_{P'} - 1}{n}\right). \tag{9}$$

Performing the configurational average we obtain

$$\mathcal{L}_{\text{eff}} \simeq \sum_{\alpha=1,r=1}^{N/2,n} \left[\tfrac{1}{2}E\left(\Psi_{(\alpha)}^{(r)}\right)^2 + \Psi_{\alpha}^{(r)}\Psi_{(\alpha+1)}^{(r)}\right] - \lambda^2 \sum_{\alpha=1}^{N/2}\left[\sum_{r=1}^{n}\left(\Psi_{(\alpha)}^{(r)}\right)^2\right]^2. \tag{10}$$

In calculating Eq. (10) we use a cumulant expansion, neglecting terms higher than Ψ^4. This approximation is equivalent with a Gaussian distribution of the masses. The parameter λ is the root-mean-square deviation of the optical frequencies normalized to the effective coupling between the cells.

Respectively, we obtain $\mathcal{L}_{\text{eff}}'$ with E' and λ':

$$E = r - \langle E_{\sigma}(\alpha)\rangle_P, \quad E' = r - \langle E_{\sigma}'(\alpha)\rangle_{P'};$$
$$\lambda = [\langle E_{\sigma}^2(\alpha)\rangle_P - \langle E_{\sigma}(\alpha)\rangle_P^2]^{1/2}, \tag{11}$$
$$\lambda' = [\langle E_{\sigma}'^2(\alpha)\rangle_{P'} - \langle E_{\sigma}'(\alpha)\rangle_{P'}^2]^{1/2}.$$

In order to analyze our results we perform a numerical calculation. We consider a simple cubic lattice in which there is a decoupling between motion in the x, y, and z directions. We assume that we have three force constants acting between nearest neighbors. K_x^x is the stretching force constant acting in the direction of propagation of the optic wave; K_y^x and K_z^x are the bending force constants and satisfy

$$(K_y^x + K_z^x)/K_x^x < 1.$$

This problem might be approximated in the case of the optical wave by only a one-dimensional problem. Each three-dimensional cell of b^3 atoms transforms to a system of b^2 chains of b atoms; K_y^x and K_z^x transform to forces acting between chains, so that $K_y^{x(\text{new})} = bK_y^x$ and $K_z^{x(\text{new})} = bK_z^x$. In the next step, we decouple the chains by approximating the intercell force constant K_x^x by $K_x^x + b(K_y^x + K_z^x)$. Such a transformation preserves the energy for long waves, but it is not valid in the case of strong disorder with correlation length comparable to b.

According to this approximation, we obtain

$$t_{\text{eff}}^{(d=3)} = t_{\text{eff}}^{(d=1)}[1 + b(K_y^x + K_z^x)/K_x^x]; \tag{12}$$

and the disorder parameter is replaced for $b = 2$ by

$$\lambda = \lambda^{(d=3)} = \lambda^{(d=1)}[1 + 2(K_y^x + K_z^x)/K_x^x]^{-1} \tag{13a}$$

and for $b = 4$ by

$$\lambda' = \lambda'^{(d=3)} = \lambda'^{(d=1)}[1 + 4(K_y^x + K_z^x)/K_x^x]^{-1}. \tag{13b}$$

We define the function $R(\lambda)$:

$$R(\lambda) = \lambda' - \lambda. \tag{14}$$

The function $R(\lambda)$ describes the transition from one-mode to two-mode behavior [using Eq. (11), we obtain for the one-dimensional case $R(\lambda) > 0$]. We can physically explain this claim as follows: The appearance of a localized mode is the condition for a two-mode behavior. For a localized mode, distant regions are uncoupled for our effective chain (the correlation function decreases exponentially). We calculate the coupling constant between distant cells as a coupling of adjacent cells in a given state of the RG transformation; increasing the size of the cell during the transformation, we obtain a decrease of $t_{\text{eff}}^{(d=3)}$, an increase of λ, and therefore $R(\lambda) > 0$. For a one-mode behavior (one type of oscillation) a long correlation exists and $t_{\text{eff}}^{(d=3)}$ decreases slowly relative to the decrease of the root-mean-square deviation of the oscillation frequencies, λ' decreases, and $R(\lambda) < 0$.

The fixed points $R(\lambda) = 0$ occur at $\lambda = 0$, $\lambda = \infty$

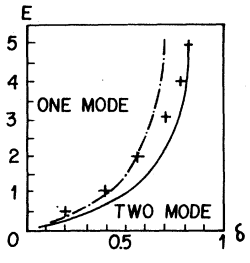

FIG. 1. Mode behavior $\epsilon = \epsilon(\delta)$ for $Z = 0.5$. Dash-dotted line, CPA; solid line, MREI; plusses, RG.

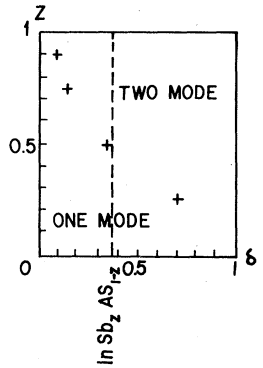

FIG. 2. Mode behavior for $Z = Z(\delta)$ for $\epsilon = 1$.

(one mode, two mode) and the unstable fixed point at $\lambda=\lambda_c\neq 0$ [$R(\lambda)>0$ for $\lambda>\lambda_c$ and $R(\lambda)<0$ for $\lambda<\lambda_c$], which describes the point of transition from one-mode to two-mode behavior.

We choose a typical[9] ratio $K_y^x/K_x^x\simeq 0.25K_z^x/K_x^x\simeq 0.25$. For a given Z and ϵ, we find λ_c for which $R(\lambda_c)=0$ and, respectively, the value of δ_c [$\lambda=\lambda(Z,\epsilon,\delta)$]. We plot a graph of ϵ vs δ ($\delta=\lambda_c$) for constant Z and a graph of Z vs δ ($\delta=\delta_c$) for constant ϵ. Figure 1 plots our results for $Z=0.5$ in comparison with those from MREI (Ref. 1) and CPA (Ref. 2). Figure 2 plots the results for $\epsilon=1$, which might explain the behavior of the crystal $InSb_zAs_{1-z}$ where $\epsilon=1.06$ and $\delta=0.38$ and which has been found to behave as one mode for $Z=0.25$ and two mode for $Z=0.85$.

We mention that our approximation might change the value of the unstable fixed point $\lambda=\lambda_c$ (to smaller λ_c) and as a result the function $Z=Z(\delta)$ (Fig. 2) appears to be shifted upwards for δ close to 1, as this approximation involves less disorder.

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[1]I. F. Chang and S. S. Mitra, Adv. Phys. 20, 359 (1971).

[2]P. N. Sen and W. M. Hartman, Phys. Rev. B 9, 367 (1973).

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

[4]D. Schmeltzer and R. Beserman, Phys. Rev. B 22, 6330 (1980).

[5]C. Herscovici and M. Fibich, to be published.

[6]M. N. Barber, Phys. Rev. 29, 1 (1977).

[7]E. Domany and S. Sarker, Phys. Rev. B 20, 4726 (1979).

[8]S. F. Edwards and M. Warner, J. Phys. A 13, 381–396 (1980).

[9]R. M. Martin, Phys. Rev. B 1, 4005 (1970).