

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 $n=0$ trick,⁴ and the recursion method⁵ have been applied. We propose here a new criterion based on the renormalization-group⁶ (RG) method, which is in agreement with experimental¹ 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 Lagrangian 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

$$\mathcal{L}_0 = \frac{1}{2} [(\epsilon_1 r - 2)x_1^2 + (r - 2)x_2^2 + 4x_1 x_2] + x_2(x_3 - x_1) + \frac{1}{2} [\dots], \quad (1)$$

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 $\Psi\sigma_2$:

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

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

The probability distribution of the eigenvalues is

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

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

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

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

$$\mathcal{L}_0 = \frac{1}{2} [(\epsilon_1 r - 2)x_1^2 + (r - 2)x_2^2 + (\epsilon_3 r - 2)x_3^2 + (r - 2)x_4^2 + 2x_1 x_2 + 2x_2 x_3 + 2x_3 x_4 + 2x_1 x_4] + x_4(x_5 - x_1) + \frac{1}{2} [\dots]. \quad (3)$$

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:

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

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

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

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

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

The probability distribution in this case is given by

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

As in the case with two atoms we obtain

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

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

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

$$t'_{\text{eff}} = \left[\left(\frac{2|ab| + 2|b|}{1 + a^2 + 2b^2} + \frac{\epsilon(1 - \delta) + \epsilon}{2(1 + \epsilon^2)^{1/2}[1 + \epsilon^2(1 - \delta)^2]^{1/2}} \right) Z^2(1 - Z)^2 + \frac{\epsilon}{(1 + \epsilon^2)} Z^4 \right. \\ \left. + \frac{\epsilon(1 - \delta)}{2[1 + \epsilon^2(1 - \delta)^2]} (1 - Z)^4 + \frac{2|b| + \epsilon(|a| + 1)}{(1 + a^2 + 2b^2)^{1/2}[2(1 + \epsilon^2)]^{1/2}} Z^3(1 - Z) \right. \\ \left. + \frac{2|b| + \epsilon(1 - \delta)(|a| + 1)}{(1 + a^2 + 2b^2)[2(1 + \epsilon^2)(1 - \delta)^2]^{1/2}} Z(1 - Z)^3 \right]. \quad (5b)$$

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

$$E_{\sigma}(\alpha) \equiv [r - v_{\sigma}(\alpha)]/t_{\text{eff}}; \quad E_{\sigma'}(\alpha) \equiv [r - v_{\sigma'}(\alpha)]/t'_{\text{eff}}. \quad (6)$$

The Lagrangians \mathcal{L} and \mathcal{L}' become

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

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

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

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

Performing the configurational average we obtain

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

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' :

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

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 .

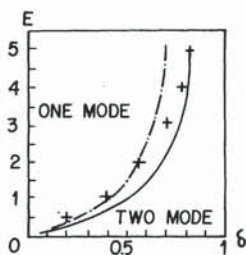


FIG. 1. Mode behavior $\epsilon = \epsilon(\delta)$ for $Z = 0.5$. Dash-dotted line, CPA; solid line, MREI; plusses, RG.

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 $\lambda = 0$, $\lambda = \infty$

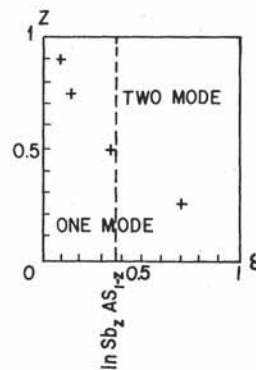


FIG. 2. Mode behavior for $Z = Z(\delta)$ for $\epsilon = 1$.

(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 typical⁹ ratio $K_y^x/K_x^x \approx 0.25K_x^x/K_x^x \approx 0.25$. 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 ($\delta = \lambda_c$) 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 $\text{InSb}_z\text{As}_{1-z}$ 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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