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Spectra of Ternary Alloys

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The vibrational frequency-distribution spectra of the random one-dimensional ternary alloy $A_c B_{1-c} C$ are accurately predicted for the first time by a statistical theory of small clusters embedded in a coherent-potential-approximation effective medium. The theory can be applied practically to real three-dimensional alloys.

By treating small clusters embedded in an effective medium (described here by the coherent-potential approximation), we are able to reproduce the "exact" numerical frequency-distribution spectra of the vibrating, mass-disordered, random, one-dimensional (1D) alloy $A_c B_{1-c} C$. The atoms of this linear ternary alloy are coupled by nearest-neighbor harmonic forces with force constants φ , and execute only longitudinal vibrations. The present theory is valid for all concentrations and is especially applicable to concentrations $0.05 \leq c \leq 0.95$ throughout the alloy regime. We believe it to be the first statistical theory to reproduce the exact numerical vibrational state densities for the 1D ternary alloy. The theory can be applied to alloys of arbitrary mass ratios m_A/m_B and m_B/m_C ; it satisfies the oscillator strength sum rule; and it correctly reproduces the exact spectra in the limits $c \rightarrow 0$ and $c \rightarrow 1$. By far the greatest virtue of this theoretical method is that it is computationally efficient, and, unlike direct numerical calculations^{1,2} and cluster gen-

eralizations of the coherent-potential approximation,³⁻⁷ can be practically applied to real 3D alloys.

The basis of the computations is the coherent-potential approximation (CPA) originally invented by Taylor,⁸ Soven,⁹ and Velický, Kirkpatrick, and Ehrenreich,¹⁰ and subsequently adapted for ternary alloys by Taylor¹¹ and by Sen and Hartmann.¹² By itself, the CPA does not give a successful quantitative description of the alloy vibrational spectra; but when a relatively small random cluster is embedded in the CPA effective medium, the resulting cluster density of states accurately mimics the density of states of the random alloy.

To our knowledge, the first authors to publish the suggestion that effective-medium and cluster methods be merged were Gonis and Garland,¹³ whose suggestion dealt only with binary alloys; however, the general idea of treating a cluster embedded in a medium has been advanced independently by numerous authors^{7,14} and we were led to the CPA as an optimal medium by our syste-

matic studies¹⁴ of various approximations for effective hosts for clusters.

The equations of motion of the infinite vibrating alloy are

$$(\underline{M}\omega^2 - \underline{\Phi})|u\rangle = 0, \quad (1)$$

where $\langle n, \alpha | u \rangle e^{-i\omega t}$ is the displacement of the α th atom in the n th unit cell, and $\alpha=1$ ($\alpha=2$) refers

to the sublattice with the A or B (C) atoms. The mass matrix is

$$\langle n, \alpha | M | n, \beta \rangle = \delta_{n,n'} \delta_{\alpha,\beta} (m_n \delta_{\alpha,1} + m_c \delta_{\alpha,2}), \quad (2)$$

where m_n is a random variable which assumes the values m_A and m_B with probability c and $1-c$.

The notation throughout the rest of the paper will thus be that the sublattice with index 1 is disordered. The force-constant matrix is expressible in terms of the Kronecker δ :

$$\langle n, \alpha | \Phi | n, \beta \rangle = \varphi [\delta_{n,n'} (3\delta_{\alpha,\beta} - 1) - \delta_{n,n-1} \delta_{\alpha,1} \delta_{\beta,2} - \delta_{n,n+1} \delta_{\alpha,2} \delta_{\beta,1}]. \quad (3)$$

In the present theory, for a fixed composition c , one selects from the disordered infinite chain a segment of N_c unit cells with a specific configuration of A and B atoms and with the ratio of the number of A atoms to the number B atoms in segment equal to c . This cluster is then embedded in an effective medium described by the Sen and Hartman¹² ternary CPA Green's function, which is defined as

$$\underline{g} = (\underline{M}_0 \omega^2 - \underline{\Phi}_0 - \underline{\Sigma} + i0)^{-1}, \quad (4)$$

where $\underline{\Sigma}$ is the self-consistently determined CPA self-energy matrix, and \underline{M}_0 and $\underline{\Phi}_0$ are the mass and force-constant matrices of any reference lattice. (Here we take the reference lattice to be BC , and recognize that the spectra are independent of this choice.¹²) For the 1D ternary, Sen and Hartmann¹² have shown that

$$\underline{\Sigma}(\omega) = \sum_n |n, 1\rangle m_B \omega^2 \bar{\sigma}(\omega) \langle n, 1|, \quad (5)$$

where $\bar{\sigma}$ satisfies the CPA self-consistency equation

$$m_B \bar{\sigma} + (m_A - m_B)c + m_B \omega^2 \bar{\sigma} (m_B \bar{\sigma} - m_B + m_A) \langle n, 1 | g | n, 1 \rangle = 0. \quad (6)$$

Equation (6), when combined with the explicit expression for $\langle n, 1 | g | n, 1 \rangle$ obtained by Sen and Hartmann,¹² forms an easily solvable cubic equation for $\bar{\sigma}$.

The Green's function for the cluster in the effective medium is

$$\underline{G} = [\underline{M}\omega^2 - \underline{\Phi} + i0]^{-1} \quad (7)$$

and satisfies Dyson's equation

$$\underline{G} = \underline{g} + \underline{g} \underline{V} \underline{G}, \quad (8)$$

where

$$\underline{V} \equiv (\underline{M}_0 - \underline{M})\omega^2 - (\underline{\Phi}_0 - \underline{\Phi}) - \underline{\Sigma}. \quad (9)$$

In what follows, we take $\underline{\Phi} = \underline{\Phi}_0$.

The primary virtue of this formalism is that, in one dimension, the resulting matrix equations are $N_c \times N_c$, the size of the cluster, and not the size of the infinite chain. These equations are solved numerically and the density of states of squared frequencies is approximated by

$$D(\omega^2) = -(\pi N_c)^{-1} \text{Im Tr}_c(\underline{M}\underline{G}), \quad (10)$$

where Tr_c means a trace over all sites of the cluster. This is then averaged over the ensemble of all possible configurations of atoms in a cluster

of N_c cells. The resulting alloy densities of states for small clusters of $N_c = 8$ unit cells are plotted in Figs. 1 and 2, where they are compared with exact numerical calculations for 20 000-atom chains by Painter.¹⁵

Figure 1 gives the frequency distribution spectrum for the case $m_B/m_A = 2$, $m_B/m_C = 0.5$, $c = 0.5$. This corresponds to the case of an alloy in the "persistence"¹⁶ regime; that is, the nearest-neighbor coupling is too weak ($|m_A - m_B| \omega^2 \gg \varphi$) to cause the AC and BC spectra to fully hybridize, and the alloy spectrum displays features clearly identifiable with individual AC and BC lattices—the middle band is the BC optical phonon and the high-frequency band is assignable to AC .

Figure 2 shows the spectrum in the case $m_B/m_A = 2$, $m_B/m_C = \frac{4}{3}$, $c = 0.5$. In this case, the alloy is in the "amalgamation"¹⁶ regime, where the A - B mass difference is so small ($|m_A - m_B| \omega^2 \ll \varphi$) that the spectra of the alloy subcomponents are lost and a hybridization spectrum appears.

Both of these spectra are for $c = 0.5$, a concentration previously inaccessible to any statistical alloy theory. As may be seen from the figures, the agreement with the exact numerical calcula-

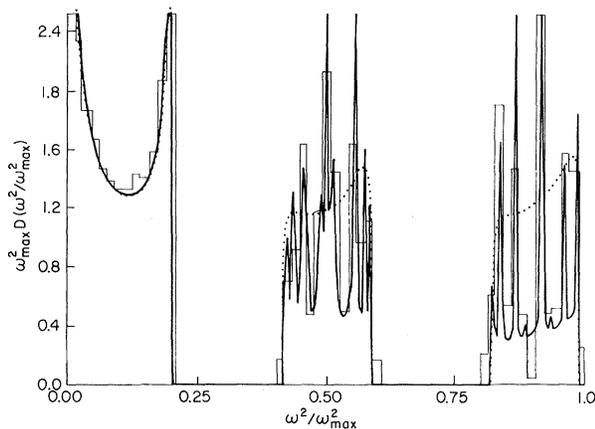


FIG. 1. Frequency-distribution spectrum for the alloy $A_c B_{1-c}$ in the "persistence" regime, $m_B/m_A=2$, $m_B/m_C=0.5$, $c=0.5$. Histogram is the exact numerical calculation of Painter for a chain of 20 000 atoms and the smooth curve is the result of the present calculation for a cluster with $N_c=8$ unit cells and a CPA effective medium. $\omega_{\max}^2=2\phi(1/m_A+1/m_C)$. The dotted line is the CPA result of Ref. 8.

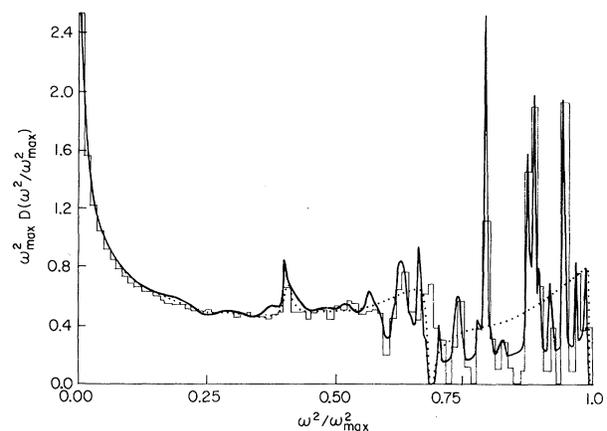


FIG. 2. Frequency-distribution spectrum for the alloy $A_c B_{1-c}$ in the "amalgamation" regime, $m_B/m_A=2$, $m_B/m_C=1/3$, $c=0.5$. Histogram is the exact numerical calculation of Painter for a chain of 20 000 atoms and the smooth curve is the result of the present calculation for a cluster with $N_c=8$ unit cells and a CPA effective medium. $\omega_{\max}^2=2\phi(1/m_A+1/m_C)$. The dotted line is the CPA results of Ref. 8.

tion is excellent in both the persistence and amalgamation limits, with the detailed features of the spectrum being reproduced for all frequencies. The CPA breaks down at very low minority-atom concentrations [order c^2 or $(1-c)^2$] and efforts to improve it by executing multisite CPA³⁻⁷ would be, especially in higher dimensions, computationally orders of magnitude more tedious than the present method even for binary alloys, since they involve integrations over the Brillouin zone and iterations of the cluster to self-consistency which are not required here.

This embedded-cluster theory can also successfully reproduce all the exact numerical spectra for the binary 1D alloy $A_c B_{1-c}$ computed by Dean¹ and by Payton and Visscher,² and can be applied to 2D and 3D binary and ternary alloys as well. A detailed discussion of the method and its application to the 1D binary alloy $A_c B_{1-c}$ and comparisons with the exact calculations of Dean¹ and Payton and Visscher² will be discussed in a later paper.¹⁴ We know of no case for which the embedded-cluster method yields spectra in poor agreement with exact numerical calculations; any such case would doubtless have to involve long-ranged resonant interactions between atoms. Moreover, the theory can be applied to electronic and magnetic excitations as well as to vibrational excitations. Therefore, we believe that the embedded-cluster theory offers a practical general solution to the alloy problem which will not only yield

spectra of random alloys but will also identify and correlate characteristic structures in those spectra with specific alloy configurations—thereby facilitating studies of nonrandom alloys.

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Direct Macroscopic Observation of Antiferromagnetic Order in Zero Applied Field

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Using superconducting-quantum-interference device magnetometry, we have made the first direct measurement of the macroscopic quadrupole field which arises from antiferromagnetic order.

Magnetization measurements are a powerful tool for investigating magnetic systems, especially ferromagnets where the sudden appearance of a large macroscopic dipole moment may be taken as direct evidence for ordering. Antiferromagnetic order on the other hand is characterized by higher-order magnetic multipoles, a quadrupole in the simplest case. Such multipoles do not give rise to strong magnetic fields in their vicinity, nor do they couple strongly to applied fields. For this reason traditional bulk magnetic measurements yield relatively little information about the antiferromagnetic state. However, the field from an antiferromagnet, though small, is not zero and one of us¹ has recently suggested that it could be measured using superconducting-quantum-interference device (SQUID) magnetometry and moreover that its spatial characteristics could be used to determine the nature of the or-

dering. In this Letter we wish to report a successful application of this idea to a simple antiferromagnet.

The principle of the method is as follows.¹ Consider a coil assembly with a small sample on its axis a distance R away. The flux from the sample which threads the coil may be shown from reciprocity² to be

$$\Phi(R) = \int_{\text{sample}} \vec{m}(\vec{r}) \cdot \vec{h}(\vec{r} + \vec{R}) d^3r, \quad (1)$$

where $\vec{m}(\vec{r})$ is the magnetization density at the point \vec{r} from the center of the sample and $\vec{h}(\vec{R} + \vec{r})$ is the field which would be produced at \vec{r} if unit current flowed in the coil. Equation (1) may be simplified without significant loss of generality by assuming the coil to have axial symmetry and m to have only an axial (z) component. If h varies only slowly over the sample, then Eq. (1) may be expanded in a Taylor series about the sample center (now at z_0) to become

$$\Phi(z_0) = \left[\int_S m_z(r) d^3r \right] h_z(z_0) + \left[\int_S z m_z(\vec{r}) d^3r \right] h_z'(z_0) + \frac{1}{2} \left[\int_S (3z^2 - r^2) m_z(\vec{r}) d^3r \right] h_z''(z_0) + \dots \quad (2)$$

Equation (2) has a simple physical interpretation. The first term is the contribution of the uniform part of m_z , i.e., of the sample's net dipole moment. The higher terms reflect the nonuniform part of m_z . Specifically the second term is given by the sample's net quadrupole moment, the third by its octupole, and so on. Furthermore, the contributions from the various multipoles may be distinguished from each other by their differing dependences on z_0 , which reflect suc-

cessively higher derivatives of h_z . These derivatives can be easily calculated from the known coil dimensions.

It is instructive to estimate some experimental magnitudes. A simple antiferromagnet (AFM) consisting of ferromagnetic planes antiferromagnetically stacked has only a quadrupole moment and yields a flux Φ_{AFM} which may be compared to the flux Φ_{FM} of the equivalent ferromagnet by the