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Evolution of discrete local levels into an impurity band in solidified inert gas solution

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The density of states $g(\omega)$ of disordered solutions of solidified inert gases are calculated using the Jacobian matrix method. The transformation of a discrete vibrational level into an impurity band at a growing concentration of light impurity atoms is investigated. It is shown that a 1-10% change in the impurity concentration leads to smearing the local discrete level into an impurity band. As this occurs, additional resonance levels appear which carry important information about the impurity-impurity and impurity-basic lattice force interactions in such solutions. © 2007 American Institute of Physics. [DOI: [10.1063/1.2746251](https://doi.org/10.1063/1.2746251)]

INTRODUCTION

It is well known that impurity atoms introduced into a crystal can cause discrete impurity levels (the so-called local oscillations) beyond the band of the quasi-continuous phonon spectrum of an ideal lattice. This occurs when the mass of the impurity atom is smaller than that of the atoms in the basic lattice or when the impurity atom-basic lattice binding is stronger than the atomic bonds in the basic lattice. The oscillations that appear are localized at the impurity atoms, and their amplitudes decrease rapidly with distance from the defect. The degree of localization is the higher the further the oscillation frequency from the upper edge of the continuous spectrum band. It is thought that the damping of the local oscillation amplitude is exponential when the distance from the defect exceeds considerably the characteristic radius of the interatomic interaction in the crystal. A systematic investigation of local oscillations was started by I. M. Lifshitz.¹⁻⁴ The conditions for the formation and the characteristics of such oscillations can be found in many monographs concerned with crystal lattice dynamics (e.g., see Refs. 5-7). At present there are numerous techniques of experimental measurement of local oscillation frequencies. Such frequencies have been obtained for many solid solutions.^{8,9} This kind of experiments provide abundant and easily obtainable (e.g., see Ref. 10) information about the parameters of defects and basic lattices.

In experiment, local frequencies can be observed in solid solutions with a finite (and small) concentration of impurity atoms in which the interaction of states at closely spaced defects is not always negligible. Because of this interaction, the localized oscillation levels can transform into impurity bands with a quasi-continuous spectrum, i.e., they alter to delocalized states.¹¹⁻¹⁵ The degree of the smearing of discrete localized levels into impurity bands is dependent not only on the impurity concentration, but also on the parameters of the defect, the basic lattice and the defect-defect interaction. It is therefore interesting to find out if the resonance character of the impurity vibrations persists at a particular matrix. If so, will the frequency of the corresponding

resonance maximum shift away from the frequency of the local oscillation induced by an isolated impurity?

At present there is a consistent theory of evolution of localized oscillations into impurity bands at low impurity concentrations¹²⁻¹⁵ when the average distance between the impurity atoms l is much larger than the atomic spacing in the basic lattice a . The power series expansion (the parameter of the expansion is $c \equiv (a/l)^3$) was obtained for the density of states (DOS) in the impurity band.

Note that at low concentrations p (commonly found as a ratio between the number of impurity atoms and the total number of atoms in the system) the l value becomes smaller than $2a$ (at $p \lesssim 2\%$ for close-packed structures, at $p \lesssim 3\%$ for a simple cubic structure, and so on). With this spacing between the impurity atoms, their effective interaction involves at least the second moments of their spectral density, which can appreciably affect (e.g., see Ref. 10) the frequencies of localized oscillations. Even at such low impurity concentrations we can observe not a discrete highly localized oscillation level but an impurity band formed by delocalized oscillations.

The evolution of discrete localized levels into impurity bands was investigated for rapidly attenuating phonons in narrow optical bands.^{16,17} However, the calculation technique proposed in Refs. 16 and 17, which was based on the Green functions and the diagram procedure, works poorly for slowly attenuating acoustic phonons.

In this study the phonon DOS of disordered solid solutions of inert gases $\text{Kr}_{1-p}\text{Pr}_p$ have been calculated numerically.

In this system the concentration p can take any value from zero to unity.¹⁸ As the concentration changes from 1 to 10%, the smearing of the local discrete level into an impurity band is attended by additional resonance levels carrying important information about the Kr-Ar and Ar-Ar force interactions in such crystals.

PHONON DENSITIES OF STATES OF SOLUTIONS OF SOLIDIFIED INERT GASES

The computation performed in this study is based on the method of Jacobian matrices (\mathcal{J} matrices) (Refs. 19-21; see

also Ref. 22). The essence of the method is the classification of vibrations, which differs from the traditional plane wave expansion. The corresponding basis $\{\vec{h}_n\}_{n=0}^{\infty}$ can be obtained through orthonormalization of the sequence

$$\{\hat{\mathcal{L}}^n \vec{h}_0\}_{n=0}^{\infty} = \vec{h}_0, \hat{\mathcal{L}} \vec{h}_0, \hat{\mathcal{L}}^2 \vec{h}_0, \dots, \hat{\mathcal{L}}^n \vec{h}_0, \dots, \quad (1)$$

which is one of possible representations of Huygens' principle. Here $\hat{\mathcal{L}}$ is the operator describing the crystal lattice vibrations:

$$\mathcal{L}_{ik}(\mathbf{r}, \mathbf{r}') = \frac{\Phi_{ik}(\mathbf{r}, \mathbf{r}')}{\sqrt{m(\mathbf{r})m(\mathbf{r}')}};$$

\mathbf{r} and \mathbf{r}' are the radius vectors of the interacting atoms; $\Phi_{ik}(\mathbf{r}, \mathbf{r}')$ is the force constant matrix describing this interaction; $m(\mathbf{r})$ and $m(\mathbf{r}')$ are the atomic masses. \vec{h}_0 is the vector in the space of renormalized atomic displacements H in which the operator $\hat{\mathcal{L}}$ acts. The vectors of this $3N$ -dimensional space (N is the number of atoms in the system) are marked with arrows to distinguish them from ordinary "three-dimensional vectors," which are shown conventionally in roman bold.

The operator $\hat{\mathcal{L}}$ in the basis $\{\vec{h}_n\}_{n=0}^{\infty}$ is represented by a three-diagonal (Jacobian) matrix (\mathcal{J} matrix). Below, a_n and b_n are used to designate the diagonal and off-diagonal matrix elements, respectively ($n \in [0; 3N \rightarrow \infty]$); the index numbering the subspaces will be omitted. This \mathcal{J} matrix has a simple spectrum, which simplifies considerably the computation of the phonon DOS. Let $\lambda \equiv \omega^2$ be the eigenvalues of the operator $\hat{\mathcal{L}}$ (squares of eigenfrequencies ω). If the band of the quasi-continuous spectrum is simply connected $\omega \in [0; \omega_m]$, the following limiting relations hold for the matrix elements a_n and b_n :

$$\lim_{n \rightarrow \infty} a_n = 2 \lim_{n \rightarrow \infty} b_n = \frac{\lambda_m}{2} \quad (\lambda_m \equiv \omega_m^2). \quad (2)$$

The arbitrary matrix elements $\mathcal{G}_{mn}(\lambda)$ of the resolvent operator $\hat{\mathcal{G}} \equiv (\lambda \hat{I} - \hat{\mathcal{L}})^{-1}$ can be represented in terms of the element $\mathcal{G}_{00}(\lambda)$ (Green function). For $m < n$ we have

$$\begin{aligned} \mathcal{G}_{mn}(\lambda) &\equiv (\vec{h}_m, \hat{\mathcal{G}}(\lambda) \vec{h}_n) = -\mathcal{P}_m(\lambda) \mathcal{Q}_n(\lambda) \\ &+ \mathcal{P}_m(\lambda) \mathcal{P}_n(\lambda) \mathcal{G}_{00}(\lambda). \end{aligned} \quad (3)$$

Here \hat{I} is the unit operator; $\mathcal{P}_n(\lambda)$ and $\mathcal{Q}_n(\lambda)$ are the polynomials to the powers n and $n-1$, respectively. They can be found in Refs. 19–22. The polynomial $\mathcal{P}_n(\lambda)$ corresponds to the determinant of the n -rank matrix of the operator $\lambda \hat{I} - \hat{\mathcal{L}}$. The polynomial $\mathcal{Q}_n(\lambda)$ is the minor of the first diagonal element of this matrix.

The Green function $\mathcal{G}(\lambda) \equiv \mathcal{G}_{00}(\lambda)$ of the system can easily be written as a continued fraction:

$$\mathcal{G}(\lambda) = \lim_{n \rightarrow \infty} \mathcal{G}_{(n)}(\lambda);$$

$$\mathcal{G}_{(n)}(\lambda) = \frac{\mathcal{Q}_n(\lambda) - b_{n-1} \mathcal{Q}_{n-1}(\lambda) \mathcal{K}_{\infty}(\lambda)}{\mathcal{P}_n(\lambda) - b_{n-1} \mathcal{P}_{n-1}(\lambda) \mathcal{K}_{\infty}(\lambda)}. \quad (4)$$

In Eq. (4) $\mathcal{K}_{\infty}(\lambda)$ is the function to which the continued fraction corresponding to the \mathcal{J} matrix whose elements are equal to their asymptotic values can be reduced. For the limiting values in Eq. (2) we have

$$\mathcal{K}_{\infty}(\lambda) = \frac{4}{\lambda_m^2} \{2\lambda - \lambda_m + 2\mathcal{Z}(\lambda) \sqrt{\lambda |\lambda - \lambda_m|}\}, \quad (5)$$

$$\mathcal{Z}(\lambda) \equiv i\Theta(\lambda)\Theta(\lambda_m - \lambda) - \Theta(\lambda - \lambda_m) \quad (6)$$

($\Theta(x)$ is the Heaviside step function).

The existence region \mathcal{D} of the imaginary part of the function $\mathcal{G}(\lambda)$, Eq. (4), determines the band of the quasi-continuous spectrum of the operator $\hat{\mathcal{L}}$ (which in general is nonsimply connected). The spectral density is estimated at $\omega \in \mathcal{D}$ to be

$$g(\omega) = \frac{1}{\pi} \text{Im } \mathcal{G}(\omega) = 2 \frac{\omega}{\pi} \text{Im } \mathcal{G}(\lambda). \quad (7)$$

The method of \mathcal{J} matrices does not include explicitly the translational symmetry of the crystal lattice and allows a straightforward computation of the spectral densities corresponding to the displacements of the atoms of the system along different crystallographical directions i . If the generating vector \vec{h}_0 is the displacement of an atom with the radius vector \mathbf{r} in the direction i , the spectral density $g_i(\omega, \mathbf{r})$ calculated by Eqs. (4)–(7) characterizes the frequency spectrum of the oscillations of this atom in this direction. The phonon DOS of a solid solution with the impurity concentration p is found as

$$\langle g(\omega, p) \rangle \equiv \frac{2\omega}{\pi N} \text{Tr } \text{Im}(\hat{I}\omega^2 - \hat{\mathcal{L}})^{-1}$$

and is a self-averaging quantity.^{12–15} It can be obtained by averaging the functions $g_i(\omega, \mathbf{r})$ over all positions of the atoms \mathbf{r} and all directions i of their displacements.

For an fcc crystal with nearest-neighbor interaction the matrix of the operator $\hat{\mathcal{L}}$ can be represented as

$$\mathcal{L}_{ik} \left(\mathbf{r}, \mathbf{r}' = \mathbf{r} + \left[\frac{a}{2}; \frac{a}{2}; 0 \right] \right) = \frac{1}{\sqrt{m(\mathbf{r})m(\mathbf{r}')}} \begin{pmatrix} \alpha & \gamma & 0 \\ \gamma & \alpha & 0 \\ 0 & 0 & \beta \end{pmatrix}. \quad (8)$$

The other matrices can be obtained through O_h symmetry operations, and the matrix $\mathcal{L}_{ik}(\mathbf{r}, \mathbf{r})$ is $(8\alpha + 4\beta) \delta_{ik} / m(\mathbf{r})$.

The force constants α , β , and γ characterizing the Kr-Kr, Kr-Ar, and Ar-Ar interactions in the $\text{Kr}_{1-p}\text{Ar}_p$ solid solution²³ were found from the elastic constants²⁴ and experimental data on the heat capacity. A random distribution of impurities was realized using a generator of pseudorandom numbers distributed uniformly in the interval (0;1). The generator operates on the basis of a multiplicative congruent method.²⁵ We calculated the phonon DOS for different concentrations of impurity atoms. At each concentration the averaging was performed over several thousand random configurations of the impurity distribution. For each configuration the DOS was found through averaging over several

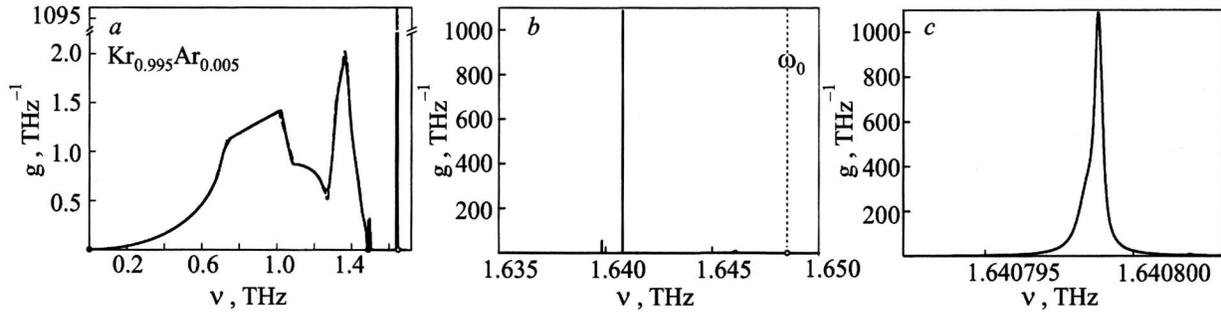


FIG. 1. Phonon density of $\text{Kr}_{0.995}\text{Ar}_{0.005}$ solid solution: *a*—the whole frequency interval; *b* and *c*—fragments of Fig. 1*a* near the local frequency. The solid curves in *a*, *b*, and *c* correspond to the function $\langle g(\omega) \rangle_{p=0.005}$; the dashed curve in fragment *a* is the phonon density of pure Kr; the thin dashed vertical straight lines (fragments *a* and *b*) are the local frequencies calculated using the “two-moment approximation” ($\nu \equiv \omega/2\pi$).

tens of spectral densities corresponding to the displacements of several tens of sequential atoms along different crystallographic directions.

The analytical properties of our calculated Jacobian matrices at $p \geq 0.1\%$ suggest unambiguously that the band of

the quasi-continuous phonon spectrum of disordered solid solutions is simply connected. The gap separating the continuous spectrum band from the local frequency in the case of an isolated impurity is filled with phonons even at limiting low concentrations of impurity atoms. The eigenfrequencies

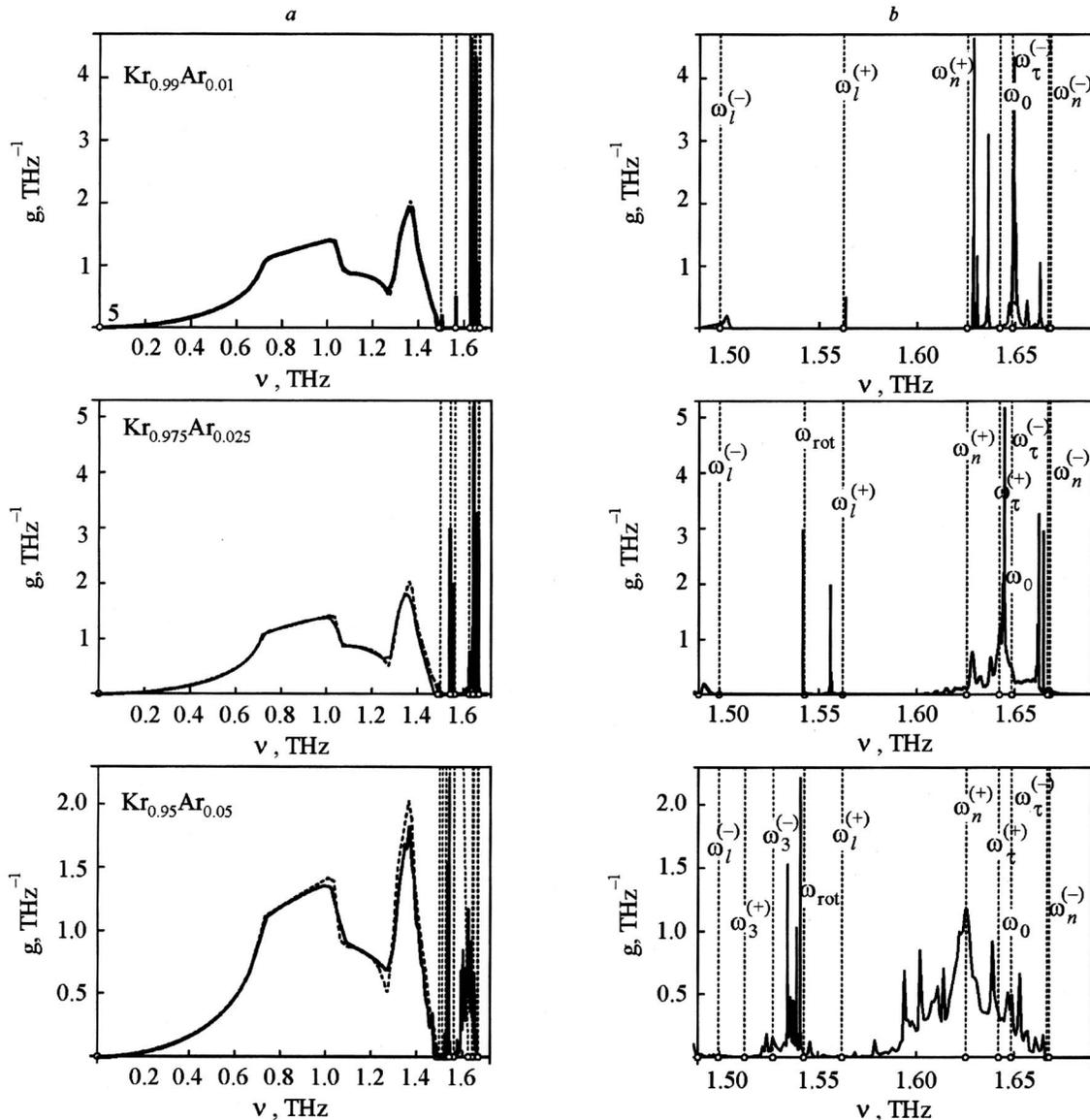


FIG. 2. Phonon densities of $\text{Kr}_{1-p}\text{Ar}_p$ solid solutions for $p=0.01, 0.025,$ and 0.05 : *a*—the whole frequency interval; *b*—beyond the quasi-continuous spectrum band, pure Kr. The solid curves (in fragments *a* and *b*) correspond to the functions $\langle g(\omega) \rangle_p$; the thin dashed vertical straight lines are the local frequencies calculated within the “two-moment approximation.” The dashed curve in fragment *a* is the phonon density of pure Kr.

are in the interval $[0, \omega_m(p)]$, where the frequency $\omega_m(p)$ is determined by the asymptotic behavior of the matrix elements.^{19–21} It is higher than the local vibration frequency corresponding to the isolated impurity with the same mass defect but lower than the so-called natural spectrum edge (e.g., see Ref. 15), i.e., lower than the highest vibration frequency of an ideal crystal lattice consisting of the atoms which we are considering as a light impurity. The latter fact is the result of the finiteness of the rank of the \mathcal{J} matrices (in our calculation it is 60), which prohibits the occurrence of an “arbitrarily large” region occupied only by impurity in the investigated configurations (covering slightly fewer than 10^6 atoms). At $p \lesssim 50\%$ the behavior of the spectral densities near $\omega_m(p)$ can be thought of as exponential attenuation, which is also suggested by the general theory of phonon spectra of disordered solid solutions.^{12–15} The simply-connectedness of the quasi-continuous spectral region in the systems analyzed permits us to calculate the Green functions and the spectral densities using their analytical approximation by a continued fraction.^{21,22} Such an approximation enables us to calculate accurately the above functions at any frequency, which is particularly important in this case when the phonon DOS spectral densities contain sharp resonance peaks.

DISCUSSION. ADDITIONAL RESONANCE LEVELS AT FINITE IMPURITY CONCENTRATIONS

Figures 1–4 show the evolution of the phonon densities $\langle g(\omega, p) \rangle$ in $\text{Kr}_{1-p}\text{Ar}_p$ solutions at growing concentration p of argon atoms. The fragments *b* in Figs. 1 and 2 are the regions of these densities corresponding to the values $\omega > \omega_m$ (ω_m is the band edge of the quasi-continuous spectrum of the ideal Kr lattice) at which these densities are significantly nonzero. Thus, the figure illustrates transformation of the local frequency into an impurity band.

The oscillations of the impurity atoms are strongly localized at $p=0.5\%$ (Fig. 1a–1c). Their frequencies are within a very narrow ($\sim 2 \times 10^6 \omega_m$) band near the frequency of the local oscillation (ω_0) caused by one isolated impurity atom. This is described with high accuracy ($\sim 25\%$) within the “two-moment approximation” proposed in Ref. 10. The local frequency calculated on the basis of this approximation is shown in Figs. 1–3 (heavy dashed curve).

It is seen in both fragments of Fig. 2 that the local level is smeared at $p=1-5\%$. The shapes of the impurity bands at these concentrations are in good agreement with the general results.^{12–15} Besides, as was mentioned in the Introduction, at $p \geq 2\%$ the average distance between the impurity atoms does not exceed the doubled atomic spacing in the lattice. In this case the influence of most impurities upon one another starts to manifest itself in the DOS at the second moment. The number of impurity pairs (the impurity atoms interacting directly with each other—the nearest neighbors in our case) becomes sufficient to show up in the phonon spectrum. For these pairs the impurity interaction is observable even at the first moment of the spectral density. It is shown in Ref. 10 that a change in the second moment of the spectral density leads to a displacement of the local level by $\pm(1-3)\%$. Such displacements are shown in Figs. 2 and 3 dashed lines) near ω_0 (not specified). A change in the first moment shifts the

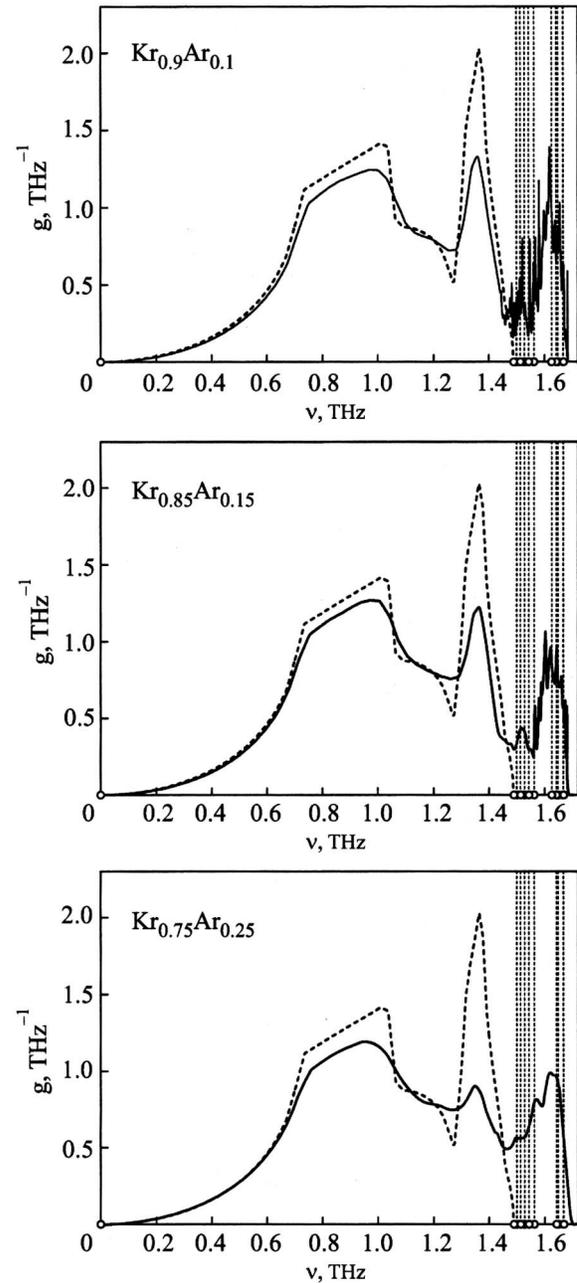


FIG. 3. Phonon densities of $\text{Kr}_{1-p}\text{Ar}_p$ solid solutions at $p=0.1, 0.15,$ and 0.25 .

local level by about $\pm(10-20)\%$. These levels ($\omega_l^{(\mp)}, \omega_\tau^{(\mp)}$, and $\omega_n^{(\mp)}$) are shown in the same figures by thin dashed lines.

The levels $\omega_l^{(\mp)}$ occur upon co- and anti-phase displacements, respectively, of two adjacent impurity atoms along the straight line connecting them. The levels $\omega_\tau^{(\mp)}$ and $\omega_n^{(\mp)}$ correspond to the displacements of two adjacent impurity atoms that are perpendicular to the above straight line.

When the adjacent atoms make up triangles, additional resonance peaks appear. The local frequencies calculated in the two-moment approximation are shown in Figs. 2 and 3 (thin dashed lines). The frequency corresponds to small rotational displacements of an equilateral triangle about the threefold axis; the frequency correlates with the displacement of the triangle as a whole and its uniform compression.

In the two-moment approximation the relation between these frequencies and the force constants characterizing the

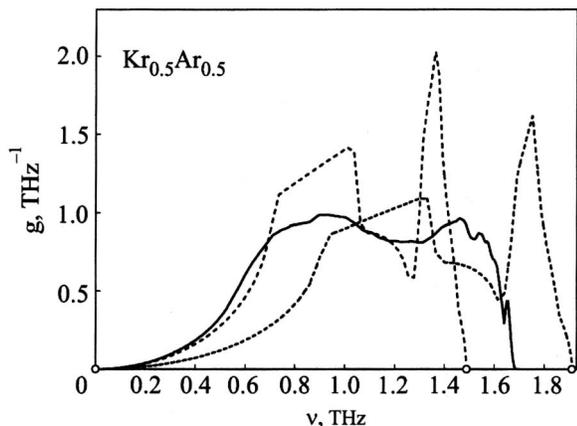


FIG. 4. Phonon density of $\text{Kr}_{0.5}\text{Ar}_{0.5}$ solid solution (solid curve). The dashed curves are the phonon densities of pure Kr and Ar.

Kr-Kr, Kr-Ar, and Ar-Ar interactions,¹⁰ which enable us to calculate the force constants from the measured frequencies of the corresponding resonance peaks.

On further growth of the concentration ($p=10\text{--}15\%$ and 25% ; Fig. 3), the impurity pairs start to interact (at the second-moment level) both with single impurity atoms and with one another. With the ratios of the mass and force constants describing the atomic interaction in the $\text{Kr}_{1-p}\text{Ar}_p$ solutions, the interaction at the level of the second moments causes the formation of a single band of the quasi-continuous spectrum at these concentrations. However, at the expression for $\omega \geq \omega_m$ the DOS has a nonanalytic form. The corresponding oscillations are quasi-localized.

Their delocalization occurs as the impurity concentration continues to increase. The phonon DOS of the $\text{Kr}_{0.5}\text{Ar}_{0.5}$ solution has no resonance peaks at $\omega > \omega_m$, and the singularity present in this frequency interval agrees with the van Hove singularity for pure Ar. This suggests that such a solution contains rather large clusters of each component, which is typical for this concentration.

CONCLUSIONS

The densities of states obtained in this study for disordered solid solutions (in particular, for solidified inert gases) with the fcc lattice and an interaction of the nearest neighbors provide at least a qualitative picture of transformation of discrete oscillation levels localized at impurity atoms into an impurity band formed by delocalized states. The main feature of the transformation is the appearance of additional impurity-induced resonance peaks at increasing impurity

concentrations. The peaks are due to the oscillations of impurity pairs and impurity clusters. The adequate description of such oscillations in the two-moment approximation¹⁰ enables one to restore in a rather simple way the parameters of the defective lattice from the measured frequencies of resonance peaks in solid solutions.

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