

## LOW TEMPERATURE PECULIARITIES OF QUASI-PARTICLE SPECTRA OF DISORDERED SOLID SOLUTIONS

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It is well known that heavy or weakly bound to the atoms of the host lattice impurities enrich the low-frequency region of the phonon spectrum and lead to a significant change in low-temperature thermodynamic and kinetic characteristics (see, for example, [1–6]). In the linear in concentration approximation the changes of the phonon spectrum due to both the heavy and light impurity atoms are well described by the Lifshitz theory of regular perturbation [7–9]. General principles of studying of quasi-particle spectra of structures containing arbitrary concentration of impurity atoms are investigated in monograph [10]. In particular, formation of the so-called quasi-local vibrations (QLV) i.e. the resonance peaks in the low-frequency phonon spectrum by heavy or weakly bound impurities, and their contribution to the low-temperature vibrational characteristics were predicted [11] and analyzed in detail theoretically (see for example, [12]) and experimentally (see, e.g., [13]). QLV are localized near the impurity atoms and their formation is very similar to the occurrence of discrete vibrational levels (local oscillations) outside the continuous spectral band of the basal lattice in the presence in crystal of light or strongly coupled impurities.

However, there is an important fundamental difference between local and quasi-local vibrations, manifested under increasing concentration of impurity atoms. Local vibrations are the poles of the Green function of the perturbed crystal, and their amplitudes decay exponentially with distance from the impurity. Being located outside of the band of quasi-continuous spectrum, these vibrations do not interact with the phonon modes of the host lattice. With an increasing content of either light or strongly coupled impurities their effect upon phonon spectrum can be defined as an expansion in the concentration (see, for example, [10]). Thus, an increasing content of light impurities leads to appearance of sharp resonant peaks in phonon spectrum at frequencies coincident with those of local vibrations which are attributed to the vibrations of the isolated impurity atom pairs and eventually, regular triangles and tetrahedra [14,15]. QLV are not the poles of Green function but common maxima in phonon density of states which do not violate its analyticity. Although these peaks are formed just by vibrations of impurity atoms, QLV interact with the phonon modes of the host lattice. Therefore, even at low enough concentrations (about few percent) of either heavy or weakly coupled impurity atoms the significant modification of the entire phonon spectrum occurs. This transformation of the entire phonon spectrum can not be described by the expansion in the concentration of impurities. As an example of the influence of the described previously

transformation of the phonon spectrum upon thermodynamic quantities we refer the “two-extreme” behaviour of the temperature dependence of the relative change in the low-temperature heat capacity of Kr-Ar solution [16, 17] (in krypton matrix interaction between argon impurities becomes weaker). This temperature dependence of heat capacity is unexplained by a superposition of contributions of isolated impurities, impurity pairs, triples and etc., without taking into account the restructuring of the entire spectrum [18]. The restructuring of the phonon spectrum of the crystal and delocalization of QLV at finite impurity concentrations were considered in [19–20] within the coherent potential approximation.

QLV usually occur in the frequency range where corresponding wavelengths of acoustic phonons of the host lattice become comparable with the average distance between the defects (the so-called disorder parameter) even at low concentrations of impurity atoms. Therefore an interaction of QLV with rapidly propagating acoustic phonons of the host lattice (so-called *propagons*) appears as the Ioffe–Regel crossover, as it is shown in [21–23], and can lead to the formation of boson peak (BP). BP is the anomalous override of low-frequency phonon density over Debye density [24]. BP were revealed in the Raman and Brillouin scattering spectra [25] as well as in experiments on inelastic neutron scattering [26] by peaks in the frequency dependence of ratio of the phonon density of states  $\nu(\omega)$  or the scattering intensity  $I(\omega)$  to the square of the frequency  $\omega$ . These peaks appear in the low-frequency region of vibrational density of states in the frequency range between 0.5 and 2 THz [27], i.e. far below the Debye frequency, while at the BP frequency a transition occurs from fast-propagated low-frequency phonons (propagons) with dispersion law close to the acoustic one, to the so-called *diffuzons*, i.e. phonons, whose propagation is hampered by scattering on localized states appearing at higher frequencies [28]. The similarity of the boson peak in disordered systems (e.g. glasses and substitution solid solutions) with the first van Hove singularity in the crystal structures is noted in [29]. BP were also observed in polymeric and metallic glasses [30–32].

However, the conditions of QLV formation and, in particular, their behaviour at a finite concentration of impurity atoms and transformation with its growth were not yet sequentially analyzed at the microscopic level. But just such an analysis allows explaining the nature of BP and provides general description for all features of low-frequency phonon spectra of real crystals and solid solutions.

The present work is devoted to analysis of the formation conditions and characteristics of QLV at the microscopic level. The evolution of QLV with increasing concentration of impurity atoms has been investigated. Within a framework of realistic crystal lattice low-frequency anomalies of the phonon spectrum, caused by both the vibrations localized on the impurities and by their scattering of the fast acoustic phonons due to vibrations of host atoms, were calculated and analyzed. Note, that BP originate from additional dispersion of

fast acoustic phonons due to their scattering on QLV, and the similarity of BP in disordered systems with van Hove singularities in regular crystal systems is analyzed.

For crystals with simple and highly symmetrical lattice the dispersion law of low-frequency phonons, i.e. phonons with frequencies lying well below the first van Hove singularity, is close to the sound dispersion law  $\omega \approx sk$  ( $\omega$  is frequency,  $k$  is a module of the wave vector  $\mathbf{k}$  and  $s$  is sound velocity) and vibrational characteristics of ideal systems are rather reasonable described within the Debye approximation. In the low-frequency region, the phonon density of states is the same as the Debye one, i.e. at  $\omega \rightarrow 0$  the function  $\nu(\omega)$  falls down as  $\omega^2$ . Therefore, the introduction of various defects into crystal can significantly enrich low-frequency region of the phonon spectrum and lead not only to quantitative but also qualitative changes in the behaviour of low-temperature vibrational characteristics.

At low impurity atoms concentrations  $p \ll 1$ , vibrational characteristics of the solid solution can be described within the linear in  $p$  approximation:

$$\tilde{\nu}(\omega) = \nu(\omega) + p \sum_i \Delta\rho_i(\omega) \quad (1)$$

The summation is performed over all cyclic subspaces with non-zero operator  $\hat{\Lambda}$  which describes perturbation of the lattice vibrations by either isolated heavy impurity or weakly coupled one,  $\Delta\rho^{(i)}(\omega)$  is the spectral density change in each of these subspaces,  $\tilde{\nu}(\omega)$  and  $\nu(\omega)$  are the phonon densities of states of solid solution and ideal crystal, respectively. If in each of the cyclic subspaces operator  $\hat{\Lambda}$  induces a regular degenerate operator, then the value  $\Delta\rho^{(i)}(\omega)$  can be calculated using the spectral shift function [8, 9]. Using the expressions obtained for this function in the J-matrix method [33–35], we obtain:

$$\Delta\rho(\omega) = -\frac{d\xi(\omega)}{d\omega} = \frac{\rho^2(\omega)}{\pi^2 \rho^2(\omega) + [S(\omega) - \text{Re}G(\omega)]} \cdot \frac{d}{d\omega} \left[ \frac{S(\omega) - \text{Re}G(\omega)}{\rho(\omega)} \right] \quad (2)$$

where  $\xi(\omega)$  is the spectral shift function [9]; the function  $S(\omega)$  describes perturbation by defect and depends on the defect parameters,  $G(\omega)$  is the local Green function of an ideal crystal.

If in any cyclic subspace the solution of equation

$$S(\omega) - \text{Re}G(\omega) = 0 \quad (3)$$

is  $\omega = \omega_k$ , then in this point vicinity the expression (2) has a resonant character.

$$-\frac{d\xi}{d\omega} = \frac{2}{\pi} \frac{\Gamma}{4(\omega - \omega_k)^2 + \Gamma^2}; \quad \Gamma \equiv \frac{\pi\rho(\omega)}{\frac{d}{d\omega}[S(\omega) - \text{Re}G(\omega)]_{\omega=\omega_k}} \quad (4)$$

Equation (3) formally coincides with the Lifshitz equation which determines (for naturally different  $S(\omega)$  function), meanings of the frequencies of discrete vibrational levels, lying outside the band of quasi-continuous spectrum of the crystal [8, 9]. However, these discrete levels in contrast to the values  $\omega_k$  are the poles of the perturbed local Green function. Green function can not have poles within the band of quasi-continuous spectrum. The possibility to determine the QLV frequencies using equation (3) is due to the fact that at low frequencies  $|\text{Re}G(\omega)| \gg \text{Im}G(\omega)$  though at  $\omega = \omega_k$  the spectral density of an ideal crystal can not be considered negligible for a lot of realistic defect-parameter values.

Let us analyze the quasi-local oscillations due to substitution impurity in FCC crystal with central nearest-neighbor interaction. Interaction of impurity with the host lattice is also considered as purely central and, therefore, as it is noted in the previous section, the perturbation caused by such impurity should be regular and degenerate. Let us consider two cases: the isotopic impurity with mass  $m$  four times higher than that of the host lattice (i.e. the mass defect is  $\varepsilon \equiv \Delta m / m = 3$ ) and the impurity atom with a mass equal to the mass of an atom of the host lattice  $\varepsilon = 0$ , though coupled to the host lattice atoms four times weaker than between each other ( $\nu \equiv \Delta\alpha / \alpha \equiv -3/4$  is the coupling defect,  $\alpha$  is the nearest neighbor interaction force constant). In the first case, operator  $\hat{\Lambda}$  induces a non-zero operator only in the cyclic subspace  $H^{(\tau^\varepsilon)} \in H$  ( $H$  is the space of atomic displacements) which is generated by displacement of the just impurity atom.

The vectors pertained to this subspace transform according to irreducible representation  $\tau_-^5$  of symmetry group of the lattice  $O_h$  (the notation of [51]). In the given subspace the spectral density of an ideal lattice coincides with its density of states. For an isotopic impurity the function  $S(\omega)$  [33] reads:

$$S_{\text{is}}(\omega) = -\frac{2}{\omega\varepsilon} \quad (5)$$

In the second case, except the subspace  $H^{(\tau^\varepsilon)}$  where the function  $S(\omega)$  reads:

$$S_w^{(\tau^\varepsilon)}(\omega) = \frac{2}{\omega} + \frac{\omega_m^2(1+\nu)}{\omega^3\nu}, \quad (6)$$

the non zero operators induced by the operator  $\hat{\Lambda}$  in cyclic subspaces are those transformed according to irreducible representations,  $\tau_+^1$ ,  $\tau_+^3$ ,  $\tau_+^4$  and  $\tau_-^4$  of the

same group  $O_h$  will be different from zero ( $\omega_m$  is the upper limit of quasi-continuous spectrum). Over all of these four subspaces  $S$ -functions coincide

$$S_w^{(\tau_1^+)}(\omega) = S_w^{(\tau_3^+)}(\omega) = S_w^{(\tau_4^+)}(\omega) = S_w^{(\tau_4^-)}(\omega) = S_w'(\omega) = \frac{16\omega}{\omega_m^2 \nu}. \quad (7)$$

For weakly bound impurity, the function  $S_w' \leq S_{\text{lim}} = 16\omega / \omega_m$ , and as shows Fig. 1, equation (3) can has no solutions within the cyclic subspaces  $H^{(\tau_1^+)}$ ,  $H^{(\tau_3^+)}$ , and  $H^{(\tau_4^+)}$ . In the subspace  $H^{(\tau_4^-)}$ , solution is possible for impurity, whose force interaction with atoms of the host lattice is weaker than the interaction of atoms of the host lattice with each other by at least 50 times. Value  $\omega_k$  will be virtually the same as the first van Hove singularity (its frequency will be labelled as  $\omega^*$ ). Therefore, for real values of parameter  $\nu$  equation (3) has a solution in the subspace  $H^{(\tau_4^-)}$  only. This solution for both cases is shown in Fig. 2.

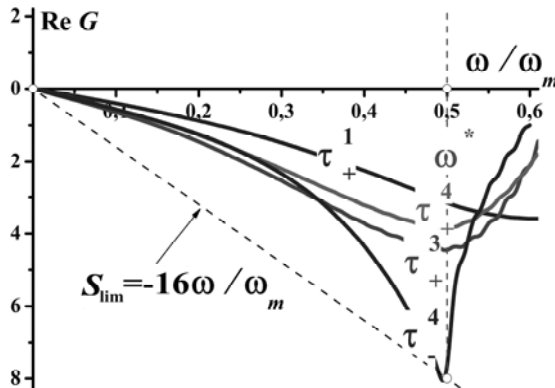


Fig. 1. Real parts of Green functions in various cyclic subspaces

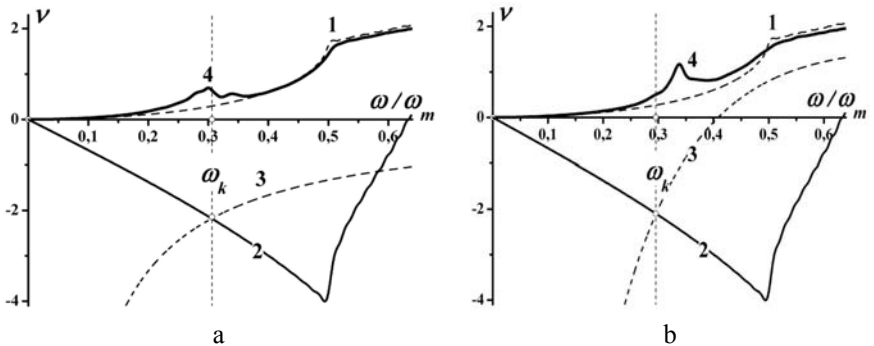


Fig. 2 Graphical solutions of equation (3) in the cyclic subspace generated by the displacement of the impurity atom: the panels *a* and *b* correspond to the heavy isotopic and weakly coupled impurities, respectively.

The real part of Green function (curves 2 in both panels) crosses the dashed curves 3, which represent the dependence (5) (the upper panel) and (6) (the bottom panel), in corresponding points  $\omega_k$ . This figure also shows the spectral densities  $\rho^{(\varepsilon)}(\omega)$  of the ideal crystal, coinciding with its phonon density of states  $\nu(\omega)$  (curves 1 (dashed)) and phonon densities of states of the corresponding solid solutions with concentration  $p = 5\%$ .

The value of the phonon density of states  $\nu(\omega_k)$  can not be considered negligible ( $\nu(\omega_k) \sim 0.1 \text{ReG}(\omega_k)$ ). Therefore, as it is seen from Fig. 2, the frequencies of the maxima in the curves 4, though they are close to frequency  $\omega_k$  but not coincident with it (especially in the case of weakly bound impurity (bottom panel). For weakly bound impurity one should expect a higher degree of localization of QLV on impurity atoms.

Modification of low-frequency density of states caused by introduction to the crystal of 5% of heavy (upper panel) and weakly bound (lower panel) impurity atoms ( $\Delta\nu(\omega) = \tilde{\nu}(\omega) - \nu(\omega)$  for curve 1) compared in Fig. 3 with the spectral density of an isolated impurity atom (curve 2 is  $0.05 \cdot \tilde{\rho}(\omega)$ )

$$\tilde{\rho}(\omega) = \frac{2\omega}{\pi} \text{Im} \left( \tilde{h}_0^{(\varepsilon)}, \left[ \omega^2 \hat{I} - \hat{L} - \hat{\Lambda} \right]^{-1} \tilde{h}_0^{(\varepsilon)} \right), \quad (8)$$

where  $\tilde{h}_0^{(\varepsilon)} \in H$  is the generating vector of the cyclic subspace  $H^{(\varepsilon)}$ ;  $\hat{L} + \hat{\Lambda}$  is perturbed operator of lattice vibrations;  $\hat{I}$  is the unit operator.

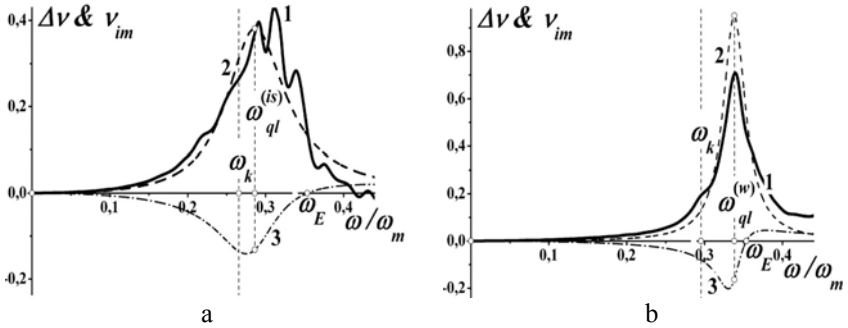


Fig. 3 Phonon density for the impurity concentration of 5% (curves 1), spectral densities of isolated impurity atoms (curves 2); spectral correlates of impurity with its nearest environment (curves 3). The panels **a** and **b** correspond to the heavy isotopic and weakly coupled impurities, respectively.

It should be noted that given spectral density has a characteristic resonance Lorentzian form similar to (4), and there are no any singularities of van Hove

type on it. The frequencies of peaks on the spectral densities  $\tilde{\rho}(\omega)$  (points  $\omega_{\text{ql}}$ ) and on the curves  $\Delta\nu(\omega)$  coincide with relatively high accuracy, especially for the case of weakly bound impurity. This indicates the strong localization of QLV on impurity atoms. Therefore the frequency  $\omega_{\text{ql}}$  can be more reasonably than  $\omega_{\text{k}}$  determined as the quasi-local frequency. Fig. 3 also represents (curves 3) the spectral correlators of displacements of impurity atoms with their first coordination sphere

$$\rho_{01}^{(\tau^{\varepsilon})}(\omega) = \frac{2\omega}{\pi} \text{Im} \left( \tilde{h}_1^{(\tau^{\varepsilon})}, \left[ \omega^2 \hat{\mathbf{I}} - \hat{\mathbf{L}} - \hat{\mathbf{\Lambda}} \right]^{-1} \tilde{h}_0^{(\tau^{\varepsilon})} \right) = \hat{\mathbf{P}}_1(\omega^2) \rho^{(\tau^{\varepsilon})}(\omega), \quad (9)$$

where  $\hat{\mathbf{P}}_1(\omega^2)$  is the polynomial defined by the recurrence relations for the  $J$ -matrix of the perturbed operator  $\hat{\mathbf{L}} + \hat{\mathbf{\Lambda}}$  [33]. Spectral correlator  $\rho_{01}(\omega)$  vanishes when  $\omega = \omega_{\text{E}}$  where  $\omega_{\text{E}}$  is the Einstein frequency of the correspondent subspace ( $\omega_{\text{E}}^2 = \int_0^{\omega_{\text{m}}} \omega^2 \nu(\omega) d\omega$ ). Thus, when  $\omega = \omega_{\text{E}}$ , a correlation with the first

coordination sphere is absent, and the closer  $\omega_{\text{ql}}$  to the  $\omega_{\text{E}}$  frequency, the degree of localization of QLV is greater. As it could be seen from Fig.3, QLV frequency for weakly bound impurity is nearly three times closer to  $\omega_{\text{E}}$  than for isotopic one, and quasi-local maximum for a weakly bound impurity has a sharper resonance form than the maximum for heavy isotopic defect.

Fig. 4 shows the evolution of the phonon densities of states of disordered solid solutions with increasing concentration of heavy isotopic impurity. We present the phonon densities of states  $\nu(\omega)$  for concentrations  $p = 0.05$ ;  $0.10$ ;  $0.25$  and  $0.5$  (solid lines in the relevant fragments of the picture). Along with these curves the dependencies  $\nu(\omega, p)$  of the perfect crystal-matrix and also of an ideal lattice, consisting of heavy ( $\varepsilon = 3$ ) atoms defined as the isotopic defect when describing the solution (thin dashed lines), are shown in each fragment as some reference point. The spectral density of the system  $\rho(\omega)$  is a self-averaging quantity (see, e.g., [10]) and can be obtained by averaging over all provisions  $\mathbf{r}$  and directions of displacement  $i$  of function  $\rho_i(\omega, \mathbf{r})$ .  $\rho_i(\omega, \mathbf{r})$  are the spectral densities in the cyclic subspaces generated by the displacements of atoms with the radius vector  $\mathbf{r}$  in the crystallographic direction  $i$ . We calculated spectral densities  $\nu(\omega, p) \equiv \langle \rho_i(\omega, \mathbf{r}) \rangle_p$  for different concentrations of randomly distributed impurity atoms. For each concentration value averaging was carried out over several thousand random configurations of the impurity distribution. For each configuration the density of states was determined by

averaging of several tens of spectral densities corresponding to displacements along different crystallographic directions of a few tens of consecutive atoms.

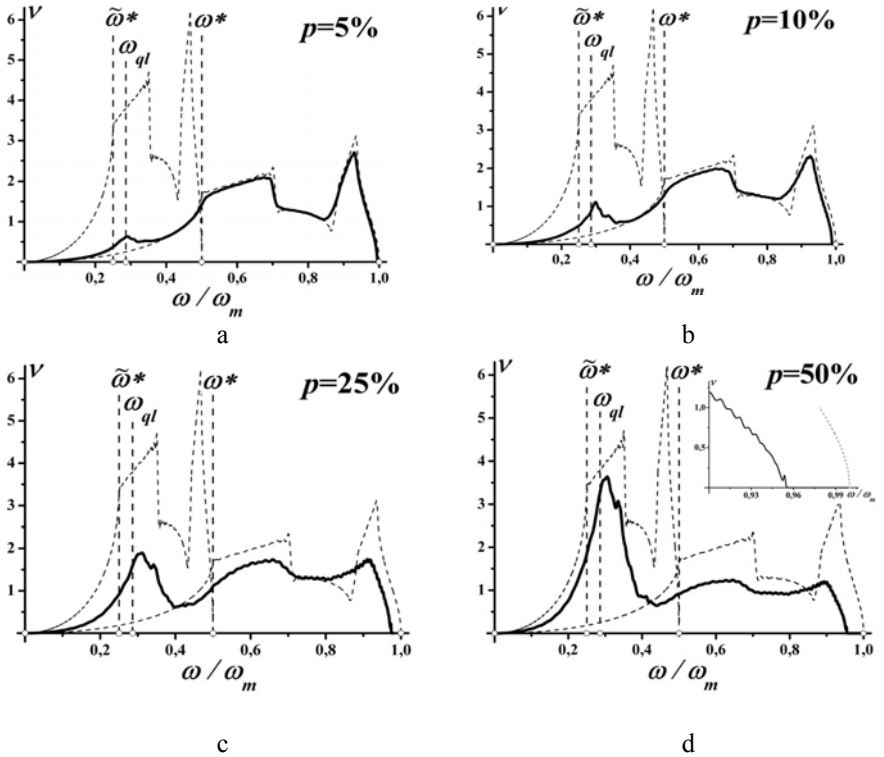


Fig. 4. Evolution of the phonon density of states of disordered solid solution with increasing concentration of heavy isotopic impurity: **a-**  $p=5\%$ ; **b-**  $p=10\%$ ; **c-**  $p=25\%$ ; **d-**  $p=50\%$ .

It should be noted here that even for  $p = 0.05$  the crystal regularity of the atoms arrangement is substantially impaired and it is impossible to use the term “*the first Brillouin zone*” for the description of the quasi-particle spectra of such a solution. At the same time the function  $\nu(\omega, p)$  near the frequency  $\omega^*$  qualitatively changes its behaviour not only when  $p = 0.05$  but also at  $p = 0.10$ , and this change is completely analogous to change of the behaviour of phonon density of states near the first van Hove feature. Let us recall that the first van Hove singularity corresponds to the change in the topology of isofrequency surfaces i.e. to the transition from closed surfaces (when  $\omega < \omega^*$ ) to open along a certain direction in reciprocal space ones (at  $\omega > \omega^*$ ) (see, e.g., [1,2]). This transition is due to touch of isofrequency to surface boundary of the first



Brillouin zone, and in the absence of the crystal translational symmetry it is impossible to talk about it.

Connection between the group velocities of acoustic phonon modes and van Hove features are analyzed more detailed in [29]. There we note that localization of phonons with certain polarization along some directions which occurs at specific frequencies is not directly related to the translational symmetry of the crystal, but will be valid also for disordered systems. For ideal crystal the frequencies of the "braking" of next phonon mode are the same for the entire sample and exhibit themselves in the behaviour of the phonon density as singularities whose form is determined by the dimension of the lattice. For structure with broken translational symmetry every "braking" mentioned earlier occurs within a certain frequency range and the correspondent singularities will be smoothed out.

Even at  $p = 0.10$  the quasi-local peak has a certain structure that remotely resembles the phonon density of states of FCC crystal, determined using a small number of moments i.e. the J-matrix of rank  $3 \div 5$  [33]. This is due to the formation of impurity clusters of appropriate size. The left slope of the peak gets parabolic shape and quasi-local frequency becomes the new boundary of propagon zone. At the same time on the phonon density it is still possible to identify high-frequency van Hove features of host lattice. With increasing concentration amount of impurity clusters increases and the boundary of propagon zone becomes more distinct. When  $p = 0.25$  this boundary is already looks as a fracture, similar to the first van Hove feature in the regular crystal. When  $p = 0.5$  the transformation of quasi-local maximum into the spectrum of the lattice of heavy atoms is substantially completed. One can clearly see two van Hove singularities: first one, separating propagons from diffuzons and another one above which phonons are propagating very slowly (locon region). In this area the curve  $\nu(\omega, 0.5)$  looks like fractal curve (similar to that obtained in [37]) for a one-dimensional solid solution). It is clearly seen in Fig. 4 (see inset on the fragment  $p = 0.5$ ) that the spectrum ends by the exponential damping of the vibrations, characteristic for disordered systems [10]. The fact that the maximum frequency in spectra shown in Fig. 4 differs from the so-called natural boundary (in this case – the maximum frequency of the host lattice) can be explained by the fact that the rank of calculated J-matrixes is finite ( $n = 76$ ) and does not allow an arbitrarily large cluster size of both the atoms of the host lattice and impurity.

Thus the condition for the existence of QLV in propagon zone is the existence of solutions of equation (3) in this frequency range. The frequency of quasi-local vibration is determined by the frequency of the maximum on the imaginary part of the local Green function of the impurity atom. Propagation of quasi-local vibrations occurs with very low velocities and can be presented as diverging waves. Scattering of quasi-plane acoustic waves on mentioned

diverging waves occurs at finite impurity concentrations  $p$  and starting from  $p \sim 10\%$  quasi-local frequency becomes the boundary of propagon zone of the phonon spectrum of solution.

So, enrichment of low-frequency part of phonon spectrum is due to not only the formation of quasi-localized states, but also to decrease of the propagation velocity of long-wavelength acoustic phonons happening due to their scattering by these states. To ensure that the results of this slowdown is clearly manifested as the boson peaks on the ratio  $\frac{v(\omega)}{\omega^2}$  or as additional singularities of Ioffe–

Regel crossovers type in the frequency range  $[0, \omega^*]$ , it is necessary to satisfy the following conditions. First, the frequencies of scattering of quasi-local vibrations have to be sufficiently low, i.e. "the power of the defect" should be sufficiently large. Secondly, the size of the defect cluster should be large enough (at least two interatomic distances) which requires a sufficiently high (15–20%) concentration of defects. The second condition means the appearance of one more parameter with dimension of length  $l \sim \frac{2\pi s_0}{\omega} > a$  ( $a$  is interatomic distance) in the system.

Parameter  $l$  plays in our case the role of the parameter of disorder. In this case, as noted in [22], the continuum approximation is not applicable even for description of the long-wavelength phonons.

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