EFFECT OF ANHARMONICITY ON THE PHONON SPECTRUM NEAR A POINT OF DEGENERACY

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The Green's function method is used to analyze the problem of the influence of anharmonicity on the phonon spectrum near a point of degeneracy. In contrast to the usual case, the influence on one another of phonon branches which are independent in the harmonic approximation requires the solution of a system of Dyson equations. Cases of essential and accidental degeneracy are treated. It is shown that in the case of accidental degeneracy the excitations that occur may have markedly different lifetimes and very different magnitudes and temperature dependence of their frequency renormalizations. In particular, the cross section for one-phonon coherent scattering of neutrons near a point of degeneracy is investigated.

1. Recently various papers[1-3] have investigated the effect of anharmonicity on the phonon spectrum and on the cross section for coherent scattering of slow neutrons. These papers tacitly assumed that all the phonon branches are strictly separated from one another. Consequently the anharmonicity resulted in an independent renormalization of frequency and appearance of damping for each individual phonon. On the other hand, the width of the peak for coherent scattering corresponding to a fixed momentum transfer κ and energy transfer ΔE was uniquely determined by the damping (the imaginary part of the corresponding polarization operator) of the phonon of the individual branch with quasimomentum $f = \kappa$ and frequency $\omega_{f,\alpha} \approx \Delta E$.

But the problem is essentially changed if the branches of the phonon spectrum intersect or merely approach close to one another. In this case the anharmonicity results in the mixing of branches that are independent in the harmonic approximation, and the resulting laws of dispersion and damping of the phonons, as well as scattering of neutrons, must be obtained by treating all the phonon branches simultaneously. Formally this corresponds to having a system of equations in place of a single Dyson equation. The solution of this problem is treated in the present paper.

2. To find the phonon spectrum we look for the retarded (advanced) Green's function $G_{\lambda\lambda'}(\omega)$, the poles of whose analytic continuation determine the laws of dispersion and damping. By definition $G_{\lambda\lambda'}(\omega)$ is the Fourier component of the function:

$$G_{\lambda\lambda'}(t-t') = -i\theta(t-t')\langle[A_\lambda(t), A_{-\lambda'}(t')]\rangle. \quad (1)$$

Here $A_\lambda = a_\lambda + a^+_{-\lambda}$, while the symbol λ denotes the set of indices f, α ($-\lambda = -f, \alpha$), where f is the quasimomentum and α labels the branch. The indices λ and λ' in (1) have the same value of f and differ only in their branch numbers.

For the Green's function (1) one can write the Dyson equations in the following form:

$$G_{\lambda\lambda'}(\omega) = G_{\lambda\lambda}^{(0)}(\omega)\delta_{\lambda\lambda'} + G_{\lambda\lambda}^{(0)}(\omega)P_{\lambda\lambda''}(\omega)G_{\lambda''\lambda'}(\omega), \quad (2)$$
$$G_{\lambda\lambda}^{(0)}(\omega) = 2\omega_\lambda / (\omega^2 - \omega_\lambda^2).$$

The polarization operator P is defined by the expression

$$P_{\lambda\lambda'}(\omega) = \frac{1}{2\hbar^2}\sum_{\lambda_1,\lambda_2} B_{-\lambda,\lambda_1,\lambda_2}B^*_{-\lambda',\lambda_1,\lambda_2}$$
$$\times\left\{\frac{n_{\lambda_1}+n_{\lambda_2}+1}{\omega-\omega_{\lambda_1}-\omega_{\lambda_2}} - \frac{n_{\lambda_1}+n_{\lambda_2}+1}{\omega+\omega_{\lambda_1}+\omega_{\lambda_2}} + 2\frac{n_{\lambda_1}-n_{\lambda_2}}{\omega+\omega_{\lambda_1}-\omega_{\lambda_2}}\right\}$$
$$+\frac{1}{\hbar}\sum_{\lambda_1} B_{-\lambda,-\lambda',\lambda_1,-\lambda_1}\left(n_{\lambda_1}+\frac{1}{2}\right),$$

where all the notation is standard.

Even for the most general form of anharmonic interaction, the polarization operator is diagonal in the index f. Thus in the general case Eq. (2) is a system of $3q$ algebraic equations where q is the number of atoms in a unit cell. Usually this equation is analyzed for $\lambda = \lambda'$ (and $\lambda'' = \lambda$). But, in treating the problem near a point of intersection of branches of the phonon spectrum, we must consider the system of equations (2).

For simplicity we shall restrict ourselves to a monatomic lattice (the extension to the more general case is trivial). Then the solution of the system (2) can be written in the form

$$G_{\alpha\alpha'}(\omega) = \frac{(-1)^{\alpha+\alpha'}M_{\alpha'\alpha}}{\Delta(\omega)}, \qquad G_{\alpha\alpha'}(\omega) = G_{\alpha'\alpha}(\omega), \quad (3)$$

$$\Delta(\omega)=\begin{vmatrix} G_{11}^{(0)-1}(\omega)-P_{11}(\omega) & -P_{12}(\omega) & -P_{13}(\omega) \\ -P_{21}(\omega) & G_{22}^{(0)-1}(\omega)-P_{22}(\omega) & -P_{23}(\omega) \\ -P_{31}(\omega) & -P_{32}(\omega) & G_{33}^{(0)-1}-P_{33}(\omega) \end{vmatrix}$$

(where $M_{\alpha\alpha'}$ is the corresponding minor, and the index f has been dropped for simplicity). We note that in the range of ω where the function $G_{\alpha\alpha'}$ is defined, the determinant of the system (2) does not vanish. Thus the system of homogeneous equations has only the trivial solution.

Now we find the poles of the analytic continuation of the function (3), as given by the solution of the equation

$$\Delta(\omega)=0. \tag{5}$$

Suppose that two phonon branches intersect for some value of f, so that $\omega_{f,1}=\omega_{f,2}=\omega_0$, and suppose that $|\omega_{f,3}-\omega_0|\gg P_{\alpha 3}$. Then, introducing the notation

$$Z=\omega^2-\omega_1^2,\ \delta=\omega_2^2-\omega_1^2,$$
$$\Pi_{\alpha\alpha'}(\omega)\ =2(\omega_\alpha\omega_{\alpha'})^{1/2}P_{\alpha\alpha'}(\omega),$$

we write the approximate solution of (5) near the point of degeneracy as

$$Z={}^1/_2(\Pi_{11}(\omega_0)+\Pi_{22}(\omega_0)+\delta)\pm[{}^1/_4(\Pi_{11}(\omega_0)+\Pi_{22}(\omega_0)+\delta)^2-(\Pi_{11}(\omega_0)\Pi_{22}(\omega_0)-\Pi_{12}(\omega_0)\Pi_{21}(\omega_0)-\Pi_{11}(\omega_0)\delta)]^{1/2}. \tag{6}$$

From the analysis of this expression it follows immediately that in general the renormalization and damping of both the excitations depend on the imaginary and real parts of the polarization operators for both branches and a fixed value of f.

Let us first look at the situation right at the point of degeneracy, i.e., when $\delta=0$. If the degeneracy is essential (i.e., caused by symmetry), $\Pi_{11}=\Pi_{22}$, and in addition $\Pi_{12}=0$. The last result is a direct consequence of the presence of an element of the symmetry group which transforms the polarization vector of one phonon into the polarization vector of the other. The degeneracy is not lifted, even though the anharmonicity results in both a shift and damping:

$$\omega'^2=\omega''^2=\omega_0+\Pi_{11}. \tag{7}$$

Much more interesting is the case of accidental degeneracy. (We note that this case occurs very frequently. Cf., for example, the recent work on analysis of the niobium spectrum[4].) Then $\Pi_{12}\neq 0$ and generally is of the same order as Π_{11}, Π_{22}. As a result the anharmonic interaction lifts the degeneracy. Assuming that Π_{12} is close in value to Π_{11}, Π_{22}, we have, approximately,

$$\omega'^2=\omega_0^2+\Pi_{11}+\Pi_{22}-(\Pi_{11}\Pi_{22}-\Pi_{12}\Pi_{21})/(\Pi_{11}+\Pi_{22}),$$
$$\omega''^2=\omega_0^2+(\Pi_{11}\Pi_{22}-\Pi_{12}\Pi_{21})/(\Pi_{11}+\Pi_{22}). \tag{8}$$

If we assume that $\Pi_{11}=\Pi_{22}$, we find from (6)

$$\omega'^2=\omega_0^2+\Pi_{11}+\Pi_{12},\qquad \omega''^2=\omega_0^2+\Pi_{11}-\Pi_{12}. \tag{8'}$$

Thus, when the degeneracy is lifted, the two excitations behave completely differently. It may happen that one of the excitations has a weak renormalization with temperature and weak damping, while the other excitation has a large renormalization and strong damping.

It is interesting to analyze the effect of a third phonon branch on the above result. Treating for simplicity the special case corresponding to (8′), we find from Eq. (5):

$$\omega'^2=\omega_0^2+\Pi_{11}+\Pi_{12}+(\Pi_{13}+\Pi_{23})^2/(\omega_3^2-\omega_0^2),$$
$$\omega''^2=\omega_0^2+\Pi_{11}-\Pi_{12}+(\Pi_{13}-\Pi_{23})^2/(\omega_3^2-\omega_0^2).$$

We see that for the anomalous second root, the effect is very much weakened, in general.

Near the point of degeneracy ($\delta\neq 0$) the detailed analysis of (5) or (6) becomes very involved, but the qualitative behavior is dictated completely by the behavior at the point of degeneracy.

Now let us consider the case where, though they do not intersect, the branches come relatively close to one another. From (5) and (6) we then have

$$\omega'^2=\omega_1^2+\Pi_{11}+\Pi_{12}\Pi_{21}/\delta,$$
$$\omega''^2=\omega_2^2+\Pi_{22}-\Pi_{12}\Pi_{21}/\delta.$$

From these expressions it follows that the approach of a branch can significantly change the renormalization and damping of the phonon belonging to the other branch, and, most importantly, may change its temperature dependence. In fact, if the value is comparable to δ (though smaller), in the classical temperature range the shift and damping will, contain a sizable quadratic term in addition to the term linear in T.

3. Let us determine the cross section for coherent scattering of slow neutrons for a momentum transfer κ close to the quasimomentum at the point of degeneracy. In the usual approximation

$$\frac{d^2\sigma(\Delta E, \varkappa)}{d\varepsilon d\Omega} = \frac{m^2 k' \varkappa^2}{8\pi^2\hbar^2 k_0}\frac{A^2}{M}e^{-W}$$
$$\times \sum_{\alpha, \alpha'} \frac{(\varkappa \mathbf{v}(\varkappa, \alpha))(\varkappa \mathbf{v}(\varkappa, \alpha'))}{[\omega(\varkappa, \alpha)\omega(\varkappa, \alpha')]^{1/2}} \frac{1}{e^{-\beta\Delta E} - 1} \operatorname{Im} G_{\alpha\alpha'}(\Delta E),$$
$$G_{\alpha\alpha'}(\Delta E) = G_{\alpha'\alpha}(\Delta E). \tag{9}$$

All the notation is standard. (The Green's function $G_{\alpha\alpha'}$ contains the index κ, which we again drop for simplicity.)

In accordance with (3), when the two branches intersect and the third branch is located sufficiently far away, we have

$$G_{11} = \frac{2\omega_1[(\Delta E)^2 - \omega_2^2 - \Pi_{22}]}{[(\Delta E)^2 - \omega'^2][(\Delta E)^2 - \omega''^2]},$$
$$G_{12} = \frac{2(\omega_1\omega_2)^{1/2}\Pi_{12}}{[(\Delta E)^2 - \omega'^2][(\Delta E)^2 - \omega''^2]}. \tag{10}$$

At a point of essential degeneracy $G_{12} = 0$, and the cross section for coherent scattering, if we consider (10) and (7), will have exactly the same peak as in the case of an isolated phonon branch.

Near a point of accidental degeneracy the cross section will have an anomalous form. In fact, now in summing (9) over α and α' one must also take into account the nondiagonal elements $G_{12} = G_{21}$, which in general are of the same order as the diagonal elements. If the imaginary part of the elements of the polarization matrix $\Pi_{\alpha\alpha'}$ is small compared to the real part, the cross section will have two close peaks with different intensities and different widths. If $\operatorname{Im}\Pi_{\alpha\alpha'} \sim \operatorname{Re}\Pi_{\alpha\alpha'}$, we get one unresolved peak which is markedly asymmetric, and this asymmetry changes drastically with temperature. (Note that if the anharmonic constant is small, all other causes of asymmetry are weak; cf. [1,3].)

These results are especially easily understood for the case where $\Pi_{11} = \Pi_{22}$. Using (8′), one easily finds the corresponding expressions for the function (10) at the point of degeneracy.

$$G_{11}(\Delta E) = \frac{\omega_0}{(\Delta E)^2 - \omega_0^2 - \Pi_{11} - \Pi_{12}} + \frac{\omega_0}{(\Delta E)^2 - \omega_0^2 - \Pi_{11} + \Pi_{12}},$$
$$G_{12}(\Delta E) = \frac{\omega_0}{(\Delta E)^2 - \omega_0^2 - \Pi_{11} - \Pi_{12}} - \frac{\omega_0}{(\Delta E)^2 - \omega_0^2 - \Pi_{11} + \Pi_{12}}.$$

Substituting these expressions in (9), we immediately get the picture described above.

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