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ABSTRACT

Simple model is presented in which two optical phonons form a resonance state with finite total momentum. In some special directions of the Brillouin zone the resonance becomes sharper as the total momentum increased.

РЕЗЮМЕ

Рассматривается простая модель, в которой два фонона образуют резонансное состояние с произвольным результирующим импульсом. Для некоторых специальных направлений в зоне Бриллюена резонанс тем острее, чем больше величина результирующего момента.

KIVONAT

Egyszerü modellt tárgyalunk, amelyben két optikai fononból egy rezonancia állapot épül fel tetszőleges eredő impulzus esetén. A Brillouin zóna néhány speciális irányait tekintve a rezonancia annál élesebb, minél nagyobb az eredő impulzus értéke.

I - INTRODUCTION

Recently M.H. Cohen and one of the authors (1) have explained the anomalous peak occurring in the Raman spectrum of diamond at the top of the two-optic-phonon continuum as the result of formation of two phonon bound state. The theory of such state has been developed in various directions (2), but it is still restricted to the case of zero total momentum, K = 0. The aim of the present paper is to discuss the main features of the bound or resonant states and hybridization with single phonons confining our attention to the neutron scattering measurements with arbitrary momentum transfer. Such hybridization occurring between one and two-phonon states has been experimentally found by J.F.Scott(3) studying the Raman spectrum of quartz. In this experiment the hybridization process has been studied by varying the temperature and thus changing the energy of the single soft phonon. We suggest now that by studying the momentum dependence of the hybridization one can make the hybridization process more spectacular. Throughout the present consideration it is assumed that the different phonons are coupled only by the anharmonic forces. Finally, it is suggested that the extra branch occurring in the recent neutron scattering data measured by Reese, Sinha, Brune and Tilford (4) on the hcp phase of solid He^4 might be explained as the result of hybridization of a single phonon with a two-optic-phonon resonance.

II - TWO PHONON BOUND STATE OR RESONANCE

The bound state may consist of two optic phonons, two acoustic phonons or phonons taken from different branches as well. Here we discuss the first two cases. Generally speaking, if the anharmonic phonon-phonon scattering is repulsive, a two phonon bound state may be formed above the energy region of the two-phonon continuum. In the case of infinite phonon lifetime, the bound state is formed if only the dimensionless fourth-order anharmonic coupling parameter g_4 is larger than a critical

$$\rho_2(E,t,\gamma,g_4') = \frac{1}{t}\,\rho_2\left(\frac{E}{t},\,1,\,\frac{\gamma}{t},\,\frac{g_4'}{t}\right) \qquad (6)$$

where is a dimensionless coupling parameter $g_4' = 1.51\, g_4\, \omega_o^2/16\varepsilon_1\, a^3$.

Thus the energy scale is contracted and the coupling is enhanced by the factor t^{-1}. It should be noted that the effective coupling strength $g_4'/t \to \infty$ as $\zeta \to 1$. This results from the complete shrinkage in $\rho_2^{(o)}$.

Let us now determine the critical value of the coupling constant g_{4c}' at which the bound state occurs just on the edge of the two phonon continuum (i.e. $E = 1$) in the limit $\Gamma = 0$. In the case $K = 0$, the critical value is $g_{4c}' = 1$. In this way, the critical value for an arbitrary K is :

$$g_{4c}'(\zeta) = t^{-1} = (\cos \zeta\pi/a)^{-1}.$$

Let us design by ζ_c the value of ζ for which $g_{4c}'(\zeta_c) = g_4$, then for $\zeta < \zeta_c$ we have the resonance in the continuum and for $\zeta > \zeta_c$ there is a bound state outside of the continuum, (see the lower half of Fig.2). The lineshape of the resonance for a finite lifetime is given in the upper part of Fig.2. We note again that for $\zeta = 1$, only a bound state is formed with binding energy $2\varepsilon_1\, g_4' / 1.51$. This calculation has been performed in an approximation where the function $\rho_2^{(o)}(E,0)$ is replaced by simple expressions which fit the van Hove singularities.

Summarizing the results, one may say, that for $g_4' > 1$, we have a bound state at arbitrary K but if $g_4' < 1$, then at small K we get a resonance which becomes a bound state at larger K at least in the direction (111).

III - HYBRIDIZATION OF A BOUND STATE WITH AN OPTIC PHONON

A bound state may be observed in the single phonon spectrum if it is hybridized with another (optic) phonon by a third-order anharmonic interaction. The theory applied here just follows Ref. 2., where the two-phonon state is coupled to an additional optic phonon assuming a coupling constant g_3 independent of the momenta. In Fig.3 is drawn the energy of the resonance state discussed before and the dispersion curve of the additional optic phonon which is given in (1) with the parameters $\tilde{\varepsilon}_1 = -1.2\,\varepsilon_1$, $\tilde{\Gamma} / \Gamma = 1.75$, $\tilde{\omega}_o - 2\omega_o = \varepsilon_1$. The dimensionless coupling parameter responsible for the hybridization is

$$g_3'^2 = g_3^2\, \tilde{\omega}_o\, g_4' / (g_4\, \varepsilon_1) \text{ and } g_3' = 0.1.$$

Thus, due to the hybridization two peaks occur in the spectrum of the additional phonon, which are shown in Fig.4. The momentum dependence of these two peaks are given in Fig.3. drawn by solid lines and the curves exhibit the typical structure characterizing the hybridization process.

value. Furthermore, if this value is slightly less than the critical one, then a strong two-phonon resonant state may be formed. The dimensionless coupling parameter g_4' is a product of the bare coupling constant g_4 and another parameter caracterizing the density of states at the top of the two phonon continuum which determines the number of the intermediate states. In the following, two simple models are briefly discussed.

a) two acoustic phonons

In most of the cases, the acoustic branch reaches its maximal value at some of the critical points of the Brillouin zone. In this region, the dispersion curve may be approximated by a parabola with an effective mass m, however, a cutt off must be applied at energy D measured from the top of the dispersion curve. If the bare coupling constant g_4 corresponding to the two-phonon scattering is taken to be independent of the momenta of the scattered phonons, the dimensionless coupling parameter depends on the total momentum of the phonon pair K only through the unrenormalized two phonon density of states $\rho_2^{(0)}(E;K)$ * for given K. The density of states decreases rapidly with increasing total momentum K ; furthermore the upper threshold decreases also (see Fig.1). The strongest binding is thus found at K=0 while it decreases quickly at larger momenta. The details will be published by one of the authors (I.T.) in another publication.

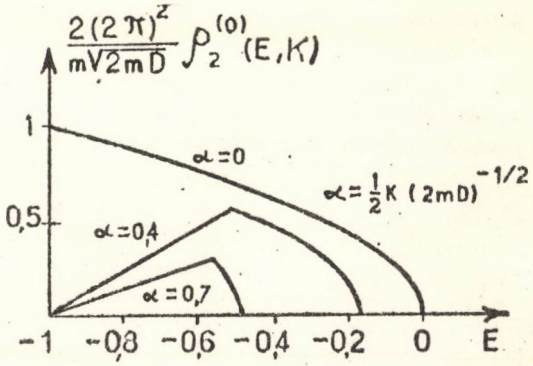

Fig.1. Two-phonon density of states without interaction for fixed values of the total momentum K expressed by the parameter α

b) two optic phonons

Nevertheless, the situation is completely reversed if one considers two optic phonons. Let us treat an oversimplified model in which
1) the Brillouin zone is cubic
2) the dispersion for the optic phonon is given by the formula

$$\omega(k) = \omega_0 + 1/3\ \varepsilon_1(\cos k_x a + \cos k_y a + \cos k_z a) \qquad (1)$$

with $\omega_0 > 3|\varepsilon_1|$. k is the momentum, a the lattice parameter.
3) The bare anharmonic coupling constant g_4 is assumed to be independent of the momenta, thus the interaction Hamiltonian can be written as $H_1 = g_4/(4!V) \int \phi\,\phi\,\phi\,\phi\, dx^3$, where ϕ is the phonon field operator defined in e.g. Ref.(2) and V is the crystal volume.

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* The energy E is measured from the top of the two-phonon spectrum.

The energy spectrum of two optic phonons exhibits a particular behavior. The energy width of the spectrum is $4\varepsilon_1$ at $K = 0$ and at finite K this width reduces drastically. Our model demonstrates this narrowing especially well for total momentum K directed in the direction (111) i.e. $K = \pi/a\ (111)\zeta$, since the energy of an arbitrary phonon pair is given by

$$\omega(\frac{K}{2} + k) + \omega(\frac{K}{2} - k) = 2\omega_o + 2/3\varepsilon_1 \cos(\zeta\pi/2)(\cos k_x a + \cos k_y a + \cos k_z a) \qquad (2)$$

thus the width of the continuous spectrum is reduced by the factor $\cos(\zeta\pi/2)$ but the total number of states is not modified(see the lower half of Fig.2.) It is important to note that

$\rho_2^{(o)} \to \delta(\omega - 2\omega_o)$ if $\zeta \to 1$.

Since the analytical form of the two phonon energy given by (2) is very simple, one can scale the case of arbitrary K to the case K=0, e.g. the unperturbed two-phonon density of states takes the form

$$\rho_2^{(o)}(E,t) = \frac{1}{t}\,\rho_2^{(o)}(1, \frac{E}{t}) \qquad [3]$$

where the following notations have been used : $t=\cos(\zeta\pi/2)$, $E = (\omega - 2\omega_o) / 2\varepsilon_1$.

In order to determine the effect of the anharmonic coupling on the two-phonon spectrum, one should solve the two-body problem or in diagrammatic language sum up the simple loop diagrams.

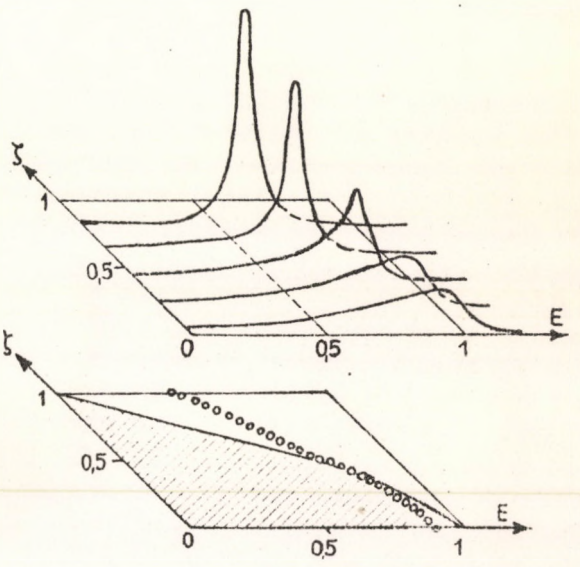

Fig.2. The two-phonon spectrum with a strong resonance at different momenta K (upper part) and the shadowed region corresponds to the unperturbed two-phonon density of states with the position of the resonance (lower part). The parameters are : $g_4' = 0.8$, $\gamma = 0.04$.

According to Ref.(2), the contribution of a single loop is

$$F(E,t) \approx 2(\frac{\omega_o}{2})^2 \int dE' \; \frac{\rho_2^{(o)}(E',t)}{E - E' + i\gamma} \qquad (4)$$

where Γ is the width of a single phonon which in the dimensionless notation reeds $\Gamma = 2\varepsilon_1\,\gamma.\rho_2(E,K)$ can be obtained in a straigthforward way

$$\rho_2(E,t,\gamma,g_4) = -\frac{1}{\omega_o^2\,\pi}\,\mathrm{Im}\left[\frac{2F(E,t)}{1 - 1/2\,g_4\,F(E,t)}\right] \qquad (5)$$

Making use of the scaling for $\rho_2^{(o)}(E,t)$ as given by (3) one gets the final form :

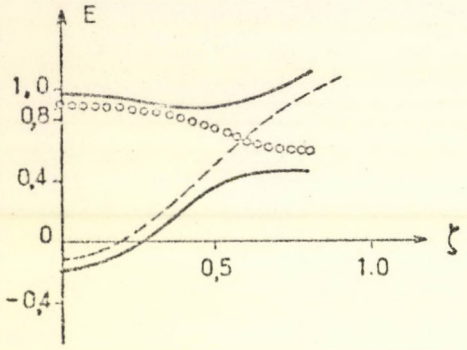

Fig.3. The momentum dependence of the energies of the peaks. Before hybridization ($g_3 = 0$) : the additional single phonon (broken line) and two-phonon bound state or resonance (dotted line), furthermore, the effect of the hybridization : two solid lines.

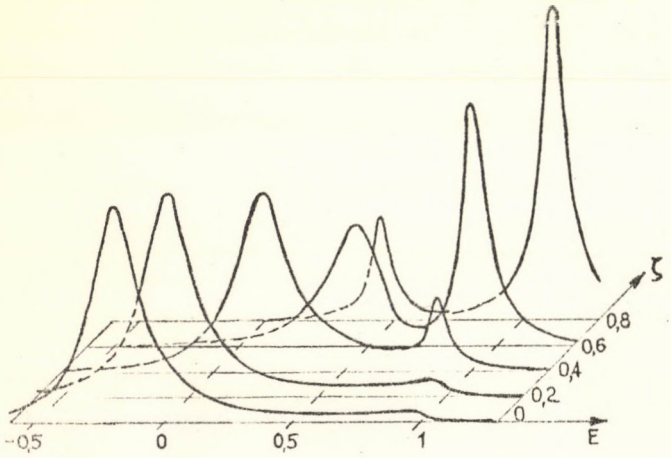

Fig.4. The spectrum of the additional single phonon at different momenta. The two peaks are due to the hybridization of the single optic phonon and the two-phonon bound state (resonance).

IV - CONCLUSION

The neutron scattering study of the hybridization process at different momenta should provide more information on the anharmonic effects in crystal dynamics. It has been suggested (5) that the additional peak occurring in neutron spectra of the hcp phase of solid He^4 (4) (6) might be due to the hybridization of an optic mode with a two phonon resonance. A careful study of its momentum dependence is necessary to decide whether the resonance is formed by the optic or two acoustic phonons.

REFERENCES

(1) M.H. COHEN and J. RUVALDS.- Phys.Rev.Lett. 23, 1378, (1969).

(2) J. RUVALDS and A. ZAWADOWSKI. - Phys.Rev. B2, 1172, (1970) and Solid State Comm. 9, 129, (1971).

(3) J.F. SCOTT. - Phys.Rev.Letters, 25, 333, (1970).

(4) R.A. REESE, S.K. SINHA, T.O. BRUN and C.R. TILFORD.- To be pubished.

(5) J. RUVALDS. - Phys.Rev. 3B, 3556 , (1971).

(6) H. HONER. - Phys.Rev. Letters, 25, 147, (1970).

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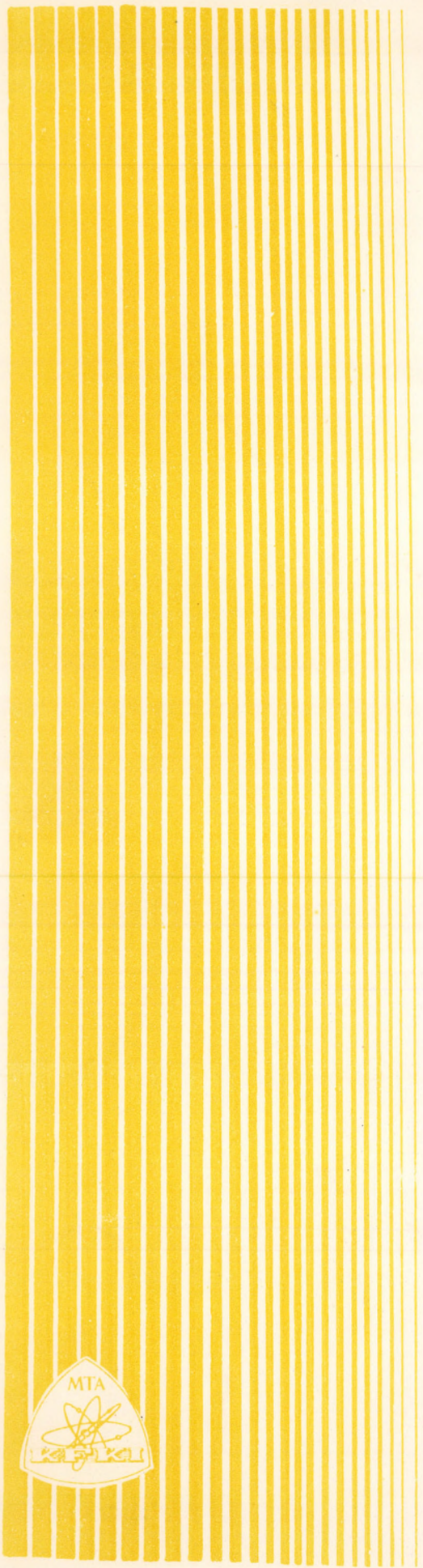

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