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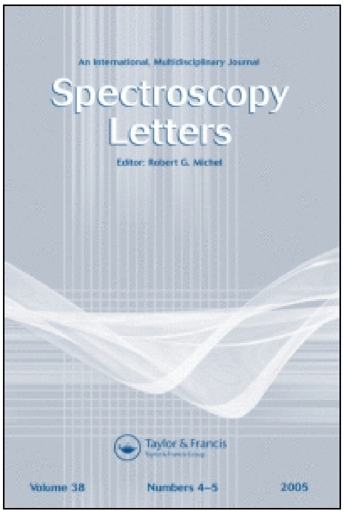

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Temperature Fermi Resonance in Crystals

I. J. Lalov[a]
[a] Faculty of Physics, University of Sofia, Sofia, Bulgaria

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TEMPERATURE FERMI RESONANCE IN CRYSTALS

Key words: Fermi resonance, sum or difference of phonon frequencies, phonon bound states, soft mode

I.J. Lalov
Faculty of Physics, University of Sofia
Sofia 1126, Bulgaria

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ABSTRACT

Some peculiarities of the crystals' vibrational spectra are studied in this work at non-zero temperatures and occasional fundamental frequency coincidence with the sum or difference of the frequencies of two other vibrations (temperature Fermi-resonance). The anharmonic interaction between the fundamental and the compound tone was shown to cause a renormalization of the spectrum also in the difference frequency region. The dielectric permittivity has been calculated in the temperature Fermi-resonance frequency region. The soft mode interaction with temperature excited difference tones in ferroelectrics has been analysed.

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1. INTRODUCTION

The Fermi-resonance phenomenon is well studied in molecular[1] and crystal[2-5] vibrational spectra; it is observed for occasional coincidence of the frequency ω_3 for a given fundamental tone with the overtone $(\omega_3 \approx 2\omega_1)$ or compound tone $(\omega_3 \approx \omega_1 + \omega_2)$ frequency. The two states with equal symmetry interact anharmonically and the mixing between the fundamental and compound tones causes renormalization of the vibrational spectrum in which typical Fermi doublets appear. The phenomenon in crystals is more complicated[3] since the fundamental tone ω_3 resonates - because of anharmonicity - with the two types of phonon states in the frequency region: (1) bound states - biphonons - that describe the movement of the two phonons in the crystal as a whole; (2) many-particle states corresponding to unbound separate propagation of the phonons in the crystal.

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Some specific peculiarities of the Fermi resonance in crystals are studied in this work at finite temperatures when the resonating phonon modes (or some of them) are temperature-excited, i.e. the phonon occupation numbers have non-zero values. The temperature amplifies the anharmonic interaction between the modes due to additional interaction between the excited real phonons[6,7]. Processes with simultaneous excitation of ω_2 phonon and absorption of real phonon with frequency ω_1 become possible at $T \neq 0$; in the frequency region $\omega_2-\omega_1$ the anharmonicity modulates the spectrum and may cause[6] one-particle state splitting (a difference biphonon). For this reason, Fermi-resonance at $T \neq 0$ - temperature Fermi-resonance - is possible also under occasional coincidence of ω_3 with the difference $\omega_2-\omega_1$, $\omega_3 \approx \omega_2-\omega_1$. This possibility is especially interesting when the resonating fundamental tone frequency ω_3 varies with temperature (soft mode, see[8,5]) or due to other reasons (for example, with variations of the concentration in mixed crystals[9]).

The influence of the coupling between one- and two-phonon states on the dielectric properties and on the infra-red crystal spectra has been investigated by Szigetti[21] in the frameworks of the perturbation theory These papers contain estimation of the infrared spectra intensities; however it is impossible to receive using perturbation theory many essential details concerning frequency dependence (for example it is impossible to investigate the presence of the bound two-phonon states[4]). In the present paper we use the more precise Green functions' method.

The subsequent text contains: (a) calculation of the cross dielectric permittivity under temperature Fermi-resonance conditions (Section 2); (b) analysis of the equation for the crystal vibrational spectrum under the same conditions (Section 3); (c) some notes on the soft

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mode interaction with two-phonon comtinuum in ferroelectrics (Section 4).

2. CROSS DIELECTRIC PERMITTIVITY AT TEMPERATURE FERMI-RESONANCE

In the theoretical study of the temperature Fermi-resonance we shall use the following model Hamiltonian:

$$\hat{H} = \sum_{\vec{k}} \hbar\omega_1(\vec{k}) b^+_{\vec{k}} b_{\vec{k}} + \sum_{\vec{k}} \hbar\omega_2(\vec{k}) c^+_{\vec{k}} c_{\vec{k}} + \sum_{\vec{k}} \hbar\omega_3(\vec{k}) a^+_{\vec{k}} a_{\vec{k}} +$$

$$+ \frac{A}{N} \sum_{\vec{K},\vec{k}_1,\vec{k}_2} (b_{\vec{k}_1} + b_{-\vec{k}_1})(b_{-\vec{K}-\vec{k}_1} + b^+_{\vec{K}+\vec{k}_1})(c_{\vec{k}_2} + c^+_{-\vec{k}_2})$$

$$(c_{\vec{K}-\vec{k}_2} + c^+_{-\vec{K}+\vec{k}_2}) + \frac{1}{\sqrt{N}} \sum_{\vec{K},\vec{k}} \Gamma(K)(a_{\vec{K}} + a^+_{-\vec{K}})(b_{\vec{k}} + b^+_{-\vec{k}})$$

$$(C_{-\vec{K}-\vec{k}} + c^+_{\vec{K}+\vec{k}}) \qquad (1)$$

where (b_k, b_k^+), (c_k, c_k^+) and (a_k, a_k^+) are the corresponding Bose operators for the interacting phonons, N is the number of unit cells in the crystal; $\Gamma(K)$ and A are anharmonic constants that express 3d and 4th power terms in the nuclear potential energy expansion in a series in normal coordinates. A similar Hamiltonian characterizes the situation when the anharmonicity is above all of intramolecular origin while the anharmonic terms generated by intermolecular interaction (see[10]) are negligible. Hamiltonian (1) is a generalization of the Hamiltonian used in[6] as well as in[2,3,11]. The last term in (1) ex-

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presses the possibility for Fermi-resonance; it is not vanishing only when the symmetry of the vibration ξ_a coincides with the symmetry of the compound tone $\xi_b \times \xi_c$, ξ_i, i = a,b,c, normal coordinates, corresponding to optical phonon.

The cross dielectric permittivity $\varepsilon(\omega,\vec{K})$ will be calculated in a dipole approximation[12-14]. For this purpose we shall find the mean value <P(t)> for the electric dipole moment (per unit cell of volume v), induced by an electromagnetic wave with electric field intensity:

$$\vec{E} = \vec{E}_o e^{i(\vec{K}.r-\omega t)} + c.c \tag{2}$$

For the cross dielectric permittivity, we obtain the expression[13,14]

$$\varepsilon_{ij}(\omega,\vec{K}) = \varepsilon_o(\delta_{ij} + \frac{i}{\varepsilon_o \omega v \hbar} \ll [\hat{P}_i(\tau),\hat{J}_j(\vec{K},0)] \gg_\omega) \tag{3}$$

where $\varepsilon_o = 8.85 \times 10^{-12}$ F/m, δ_{ij} is the Kronecker symbol, $\hat{P}$ is the electric dipole moment operator for one unit cell in the crystal:

$$\hat{P} = \sum_\mu e_\mu \hat{r}_\mu \tag{4}$$

The summation in (4) runs over all charges e_μ in the unit cell and $\hat{r}_\mu$ is the charge coordinate operator. The operator $\hat{J}$,K) in a dipole approximation has the following form[13,14]:

$$\hat{J}(K) = \sum_\beta \frac{e_\beta}{m_\beta} \hat{J}_\beta e^{-i\vec{K}.\vec{r}_\beta^{(0)}} \tag{5}$$

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The quantities m_β and $\hat{J}_\beta$ are mass and momentum operator and $r_\beta^{(0)}$ is the equilibrium position of charge β; the summation in (5) extends over all charges in the crystal. The quantity $\langle\ldots\rangle_\omega$ in (3) represents a Fourier-component by time for the following retarding Green function

$$\langle[\hat{P}_i(\tau),\hat{J}_j(\vec{K},0)]\rangle = -i\Theta(\tau)\langle[\hat{P}_i(\tau),\hat{J}_j(\vec{K},0)]\rangle \tag{6}$$

In the second quantization representation, the operators $\hat{P}$ and $\hat{J}(K)$ have been calculated in[13,14] for a fundamental tone of the vibration ω_3 and in[6,15] for the compound tones $\omega_2 \pm \omega_1$. Under Fermi resonance the operators P and $\hat{J}(K)$ represent sums of the corresponding operators for the fundamental and compound tones:

$$\hat{p} = \sum_{K}[\frac{\vec{p}_3}{\sqrt{N}}(a_{\vec{K}}+a^{+}_{-\vec{K}}) + \frac{\vec{p}_2}{N}\sum_{\vec{k}}(b_{\vec{k}}+b^{+}_{-\vec{k}})(c_{\vec{K}-\vec{k}}+c_{\vec{k}-\vec{K}})] \tag{7}$$

$$\begin{aligned}\hat{J}(\vec{K}) = {} & i\omega_3(\vec{K})\sqrt{N}\,\vec{p}_3(a^{+}_{\vec{K}}-a_{-\vec{K}}) + \\ & + i\vec{p}_2\sum_{\vec{k}}[\omega_1(\vec{k})(b^{+}_{-\vec{k}}-b_{\vec{k}})(c^{+}_{-\vec{K}+\vec{k}}+c_{\vec{K}-\vec{k}}) + \\ & + \omega_2(\vec{K}-\vec{k})(b^{+}_{-\vec{k}}+b_{\vec{k}})(c^{+}_{-\vec{K}+\vec{k}}-c_{\vec{K}-\vec{k}})]\end{aligned} \tag{8}$$

In (7) and (8) the possible dependence of the vectors $\vec{p}_2,\vec{p}_3$ on the wave vectors (K,k) has been neglected as weak[6]. Due to the equal symmetry of the fundamental tone and the compound tone, the matrix elements of the electric dipole moment $\vec{p}_2,\vec{p}_3$ are colinear (if the correspon-

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ding nondegenerate representation ξ_3 allows only one direction of the non-zero polar vector):

$$\vec{p}_2 = \lambda\vec{p}_3 \tag{9}$$

The quantity λ is usually lower and even considerably lower, than unity and it expresses the electro-optical anharmonicity.

To calculate ε we need the following Fourier components of retarding Green functions, see formulae (6) to (8):

$$g_1(\omega,\vec{K}) = \langle\langle[(a_K(\tau) + a^+_{-K}(\tau), a^+_K(0) - a_{-K}(0)]\rangle\rangle_\omega \tag{10a}$$

$$g_2(\omega,\vec{K}) = \langle\langle[\sum_{\vec{k}}(b_{\vec{k}}(\tau)+b^+_{-\vec{k}}(\tau))(c_{\vec{K}-\vec{k}}(\tau)+c^+_{\vec{k}-\vec{K}}(\tau)), \\ a^+_{\vec{K}}(0) - a_{-\vec{K}}(0)]\rangle\rangle_\omega \tag{10b}$$

and the corresponding functions g_3 and g_4 in which the first operators (τ) in the correlators are the same as in g_1 and g_2 but the second operator $\tau = 0$ has been substituted by the second addend in (8). The Green functions are calculated by a standard procedure with time differentiation and subsequent decoupling of the chain of equations for the Green functions^6. The next correlations are used:

$$\langle(b_{k_1}+b^+_{-k_1})(b_{k_2}\pm b^+_{-k_2})\rangle \approx \delta_{k\ ,-k_2}\{n^b(k_2)\pm[n^b(k_2)+1]\} \tag{11}$$

where $n^{b,c}(k) = \{\exp[\hbar\omega_{1,2}(k)/(kT)] - 1\}^{-1}$ are the Planck occupation numbers of the phonon modes. After

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decoupling of the chain, the set of equations for the Fourier components of functions (10) is solved exactly producing

$$g_1(\omega,K) = \frac{2\omega}{\omega^2-\omega_3^2(K)} \cdot \{1 + \frac{4\Gamma^2(K)\omega_3(K)\Delta_o^{-1}(\omega,K)}{\omega^2-\omega_3^2(K)} \cdot$$

$$\cdot [T_1(\omega,K) + T_2(\omega,K)]\} \qquad (12a)$$

$$g_2(\omega,K) = \frac{4\omega\Gamma(K)}{\omega^2-\omega_3^2(K)} \Delta_o^{-1}(\omega,K) \cdot [T_1(\omega,K)+T_2(\omega,K)] \qquad (12b)$$

$$g_3(\omega,K) = \omega_3(K)g_2(\omega,K) \qquad (13a)$$

$$g_4(\omega,K) = 2\omega\Delta_o^{-1}(\omega,K) \cdot [T_1(\omega,K) + T_2(\omega,K)] \qquad (13b)$$

where

$$\Delta_o(\omega,K) = 1 - 4(A + \frac{\Gamma^2(K)\omega_3(K)}{\omega^2-\omega_3^2(K)})[T_1(\omega,K)+T_2(\omega,K)] \qquad (14)$$

$$T_1(\omega,K) = \frac{1}{N}\sum_k \frac{[\omega_2(K-k)+\omega_1(k)][1+n^b(k)+n^c(K-k)]}{\omega^2-[\omega_2(K-k)+\omega_1(k)]^2} \qquad (15)$$

$$T_2(\omega,K) = \frac{1}{N}\sum_k \frac{[\omega_2(K+k)-\omega_1(k)] \cdot [n^b(k)-n^c(K+k)]}{\omega^2-[\omega_2(K+k)-\omega_1(k)]^2} \qquad (16)$$

Using formulae (3), (7), (8), (12), (13) we obtain the following expression for the tensor $\varepsilon(\omega,K)$

$$\varepsilon_{ij}(\omega,K)/\varepsilon_o = \delta_{ij} - \frac{2}{\varepsilon_o \hbar v} \{p_3^i p_3^j \frac{\omega_3(K)}{\omega^2-\omega_3^2(K)} +$$

$$+ \frac{T_1(\omega,K)+T_2(\omega,K)}{\Delta_o(\omega,K)} \cdot (p_2^i + \frac{2\Gamma(K)\omega_3(K)}{\omega^2-\omega_3^2(K)} p_3^i) .$$

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$$\cdot\ (p_2^j + \frac{2\Gamma(K)\omega_3(K)}{\omega^2-\omega_3^2(K)}\, p_3^j)\} \qquad (17)$$

When eqn (9) holds, formula (17) is simplified:

$$\varepsilon_{ij}(\omega,K)/\varepsilon_o = \delta_{ij} - \frac{2p_3^i p_3^j}{\varepsilon_o \hbar V}\{\frac{\omega_3(K)}{\omega^2-\omega_3^2(K)} + \frac{T_1(\omega,K)+T_2(\omega,K)}{\Delta_o(\omega,K)}\cdot[\lambda + \frac{2\Gamma(K)\omega_3(K)}{\omega^2-\omega_3^2(K)}]^2\} \qquad (18)$$

The constant $\Gamma(K)$ is particularly important for the manifestation of Fermi-resonance: if $\Gamma(K) = 0$, the dielectric permittivity has resonances in separate, independent regions $\omega \approx \omega_3(K)$ and $\omega \approx \omega_2(k_2) \pm \omega_1(k_1)$. If $\Gamma(K) \neq 0$ the resonances in the compound tone region are amplified (see last factor in (18)). The factor $[\lambda + 2\Gamma\omega_3/(\omega^2-\omega_3^2)]^2$ expresses in the case considered the phenomenon Fano antiresonance[16]: interaction of a discrete level $\omega_3(K)$ with continuum $\omega_2(k_2) \pm \omega_1(k_1)$. This factor may vanish if the two addends are equal in absolute values but with opposite signs (destructive interference between one- and two-phonon contribution to dielectric permittivity).

Since the imaginary part of the quantity ε determines the frequency dependence of the absorption coefficient, the Fermi-resonance intensity redistribution is manifested also in the absorption spectra[10,17]. Unlike the case $T = 0$, studied in[10], in our case Fermi reso-

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nance will appear also in the difference frequency region $\omega \approx \omega_2 - \omega_1$ ($T_2 \neq 0$ at $T \neq 0$).

3. VIBRATIONAL SPECTRUM AT TEMPERATURE FERMI RESONANCE

The vibrational spectrum is defined be the Green function poles, i.e. by the equation $\Delta_0(\omega,K) = 0$. A similar spectrum has been analysed in[3,6] and we shall point out only some peculiarities.

The effective interaction between b- and c-phonons is expressed by the anharmonicity factor

$$A'(\omega,K) = A + \frac{\Gamma^2(K)\omega_3(K)}{\omega^2-\omega_3^2(K)} \qquad (19)$$

While usually $A < 0$, the second addent changes its sign depending on the frequency ω and the following cases are possible: $A'(\omega,K) < 0$ (attraction between b- and c-phonons) and $A'(\omega,K) > 0$ (repulsion). The two continuums $\omega_1(\vec{k}_1)+\omega_2(\vec{k}_2)$ and $\omega_2(\vec{k}_2)-\omega_1(\vec{k}_1)$ form quasi-continuous bands called many-particle bands (MP bands). The discrete levels may split and they correspond to one-particle bound states - biphonons. These levels may exist above the corresponding MP band (at $A'(\omega,K) > 0$) and below it (at $A' < 0$).

The vibrational spectrum may be more suitably analysed by rewriting equarion $\Delta_0(\omega,K) = 0$ into the following form (at $\omega \approx \omega_3(K)$).

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$$F(\omega,T) = \frac{2.[T_1(\omega,K)+T_2(\omega,K)]}{1-4A[T_1(\omega,K)+T_2(\omega,K)]} = \frac{\omega-\omega_3(K)}{\Gamma^2(K)} \qquad (20)$$

Both side of eqn (20) may be temperature dependent: the left side via the occupation numbers n^b, n^c and the right hand side - with temperature variations of ω_3. In this Section we suppose that the frequency $\omega_3(K)$ is located near a given many-particle band, or $\omega_1(k_1)+\omega_2(k_2)$ or $\omega_2(k_2)-\omega_1(k_1)$ and varies weakly with temperature. In this case the temperature dependence for the spectrum is correlated with the function $F(\omega,T)$ and only one of the quantities T_1 or T_2 is resonant. Thus, if $\omega \approx \omega_3(K) \approx$ $\approx \omega_2(k_2)-\omega_1(k_1)$, resonant would be the sum T_2, and T_1 would be almost ω-independent. The temperature Fermi-resonance considered here is strongly expressed namely in the $\omega_3(K)$ frequency region. The vibrational spectrum in the second region (for example $\omega' \approx \omega_1(k_1)+\omega_2(k_2)$) located far from $\omega_3(K)$ would be practically unaffected by the Fermi resonance and would experience only the anharmonic interaction via the constant A.

Equation (20) has different solutions depending on the values of this constant A[17,18]:

(1) for weak anharmonivity $A \approx 0$ the denominator on the left side of (20) is not vanishing for any ω value (Fig. 1). It would be noted that outside the regions $\omega_2(k_2)\pm\omega_1(k_1)$ the sums T_1 and T_2 increase in absolute value with the temperature due to the increasing occupa-

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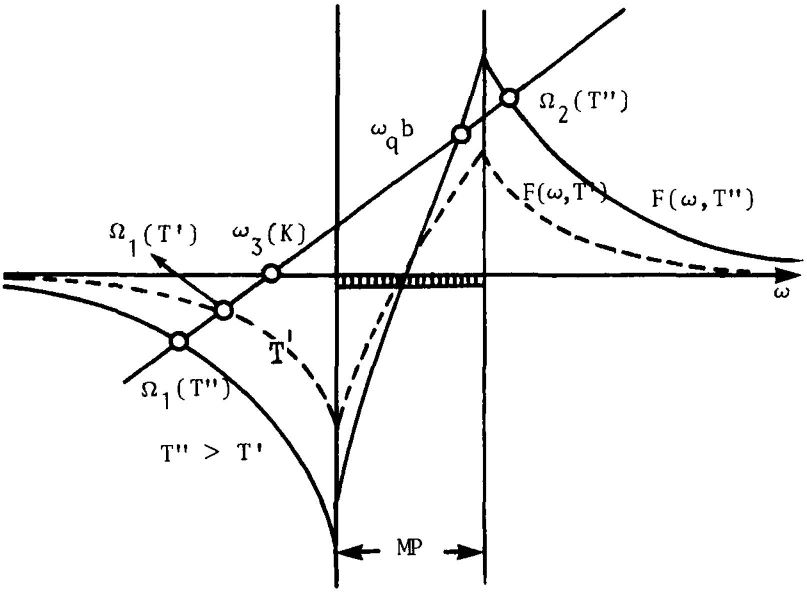

FIG. 1. Graphic solution of the equation (20), small anharmonicity case ($A \approx 0$). The curve $F(\omega,T)$ is the plot for left side of (20), the line throught $\omega_3(K)$ point is right side plot; MP - many - particle band; ω_1, ω_2 - one - particle levels below and above the band MP; ω_{qb} - quasi-bound level.

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tion numbers in the numerators. The one-particle level $\Omega_1(T)$ is removed away from the MP band with increasing temperature; however a second one-particle level $\Omega_2(T)$ may appear above this band. The solutions Ω_{qb} of (20) lying inside the MP band correspond to quasi-bound, decaying states.

(2) for large anharmonicity A there appears a pole $\omega_1(T)$ of the function $F(\omega,T)$, see Fig. 2. This pole is temperature-dependent[6] its separation from the MP band

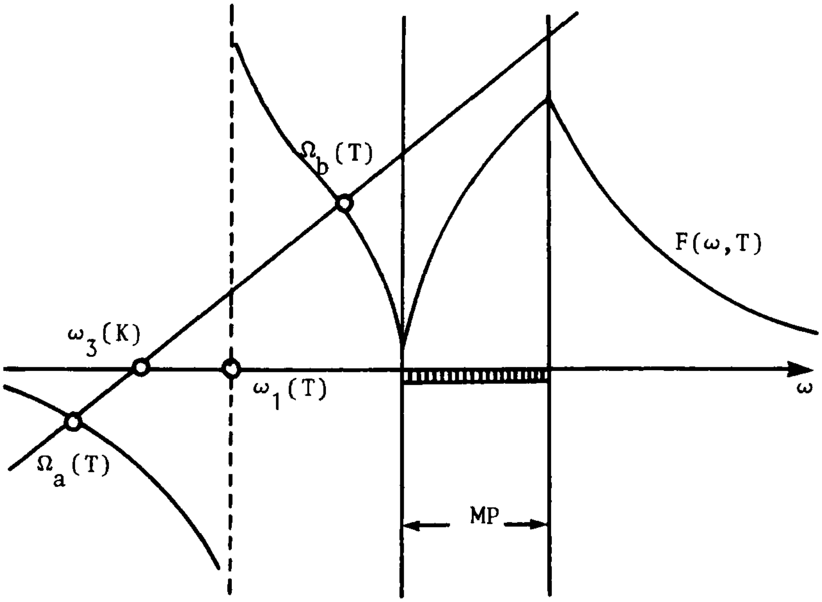

FIG. 2. Graphic solution of (20), strong anharmonicity case; $\omega_1(T)$ - the pole of $F(\omega,T)$, for the other symbols see fig. 1. Two one-particle levels - ω_a, ω_b - appear below MP.

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increasing with temperature. For large values of the Fermi resonance constant $\Gamma(K)$ a Fermi doublet of one-particle levels appears, one level being below and another above the MP band, moving away from this band with increasing T. If however the Fermi resonance is more weakly expressed (intermediate values of the constant $\Gamma(K)$, Fig. 2), the two one-particle levels Ω_a and Ω_b may lie below the MP band.

The actual picture for the spectrum may differ in details from the picture depicted here. Thus the vibrational spectrum would change if the level $\omega_3(K)$ lies above the MP band (for different cases, see[3,16,17]). At Fermi resonance in the sum region, $\omega_3 \approx \omega_1+\omega_2$ such a picture characterizes the two-phonon region even at T = 0. Fermi resonance in the difference region $\omega_3 \approx \omega_2-\omega_1$ would not appear at T = 0. The sum T_2 increases and the difference spectrum is intensified with increasing temperature: the MP band appears and consecutively also the one-particle states from Fig. 1. At even higher temperatures and considerable anharmonicity A, the picture in Fig. 2 may be observed in the difference spectrum.

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4. SOFT MODE AND FERMI RESONANCE

As known when the temperature approaches the ferroelectric phase transition temperature, $T \to T_C$, $T < T_C$, the soft mode frequency decreases and tends to zero. It falls within the frequency region lower than the fundamental optical vibrations where the differences from their frequencies are however found. If some of the vibrations are temperature excited, the considered temperature Fermi resonance generates peculiarities for the infrared and Raman spectra. It should born in mind that the soft mode is dipole active and for this reason our calculation of ε^{l} is in this case actual.

The analysis of the vibrational spectrum is once again based on eqn (20); we have to suppose that the

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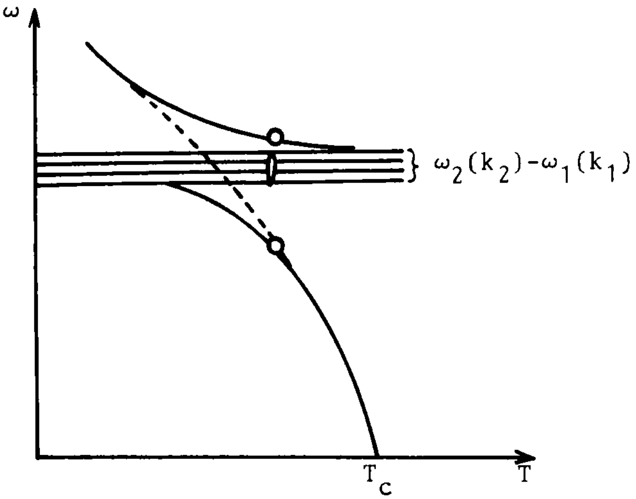

FIG. 3. Fermi-doublet in the difference frequiency region near ferroelectric phase transition, T_c - transition temperature. Dashed curve - soft mode behaviour without Fermi-resonance.

frequency $\omega_3(K,T)$ decreases with increasing temperature, i.e. it approaches from above the MP bands of Fig. 1-2, passes these bands and moves away into the low-frequency region. The passage of the soft mode through the sum and difference MP continuums causes splitting (Fig. 3), Fermi doublets and eventually Fermi triplets appearing (two one-particle lavels and one MP band). Similar spectra have been observed many times (first in quartz and ALPO$_4$[8]). The approach of the soft mode to the MP band enhances the corresponding spectral line intensity (in formula (18) $[\lambda + 2\Gamma\omega_3/(\omega^2-\omega_3^2)]^2$ the second addend in the factor may considerably exceed λ if $\omega \approx \omega_3(K,T)$.

At Fermi resonance among the soft mode and several continuums in the low-frequency region, the spectrum acquires even a quasi-continuous nature as if no separate lines are excited but whole frequency regions ($LiNBO_3$, $PB_5Ge_3O_5$, see[5]).

5. CONCLUSION

In this work, the interaction between fundamental vibrations and compound vibration equalling the sum or difference of the frequencies for two different vibrations, have been studied. The temperature excitation of some phonon modes amplifies the interaction between the quasi-particles in the crystal and provides qualitatively new possibilities: a fundamental tone interacting with a difference tone and anharmonic renormalization of the difference spectrum. Our results about the renormalization of the vibrational spectrum and intensity redistribution at temperature Fermi resonance are qualitatively confirmed[5] by experiments on the soft mode temperature behaviour. These experiments raise the problem of analytical and numerical computation of two-phonon spectra in concrete materials, see[19].

Naturally, the temperature excitation of some phonon modes amplifies their effect on the phonon line width. Some experimental data[20] raise important problems about the simultaneous influence of anharmonicity and tempe-

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rature on the shape and width of spectral lines in two-phonon spectra, which will be treated in a subsequent publication.

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