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ON THE PROBLEM OF NEUTRON SPECTROSCOPY OF PARAMETRICALLY NONEQUILIBRIUM QUASIPARTICLES IN SOLIDS.

I. Neutron Inelastic Scattering on Polaritons in the Parametric Resonance Conditions

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1. INTRODUCTION

The possibility of investigation of dynamical properties of solids by means of neutron inelastic scattering experiments is well proved at present, and the neutron spectroscopy as an independent direction in the experimental study of condensed matter in general and of crystalline solids in particular became traditional and develops intensively together with the optical spectroscopy of solids. Considerable results have been obtained both in experimental and theoretical investigations of a great number of solids in equilibrium state (see, for example, refs.[1-3]), and the advantage of the neutron inelastic scattering spectroscopy has been proved to be indubitable in many cases.

As far as the neutron spectroscopy of quasiequilibrium and nonequilibrium quasiparticles in solids is concerned, the situation is not the same here. Although this topic is already attracting a lot of attention of the theorists (see, as examples, refs.[2,4-8]), it still remains a new one in the experimental aspect. The main difficulty here is connected with too small concentrations of quasiequilibrium or nonequilibrium quasiparticles even created by rather high-power external sources (such as laser fields, heat pulses, etc....). This circumstance makes the corresponding scattering cross sections too small to be measured in experiments[4]. Nevertheless, it is of present interest to pose the question not only about theoretical but also experimental investigations in this direction since more and more high-flux neutron sources become accessible today[9]. It is worth to note here that the neutron inelastic scattering spectroscopy, being free of the selection rules, can give in many cases essentially more rich information about the quasiparticle structure as compared with the traditional optical Raman scattering methods.

In the light of the present discussion, the use of the parametric resonance phenomenon in solids seems to be a good method for solving the small concentration problem in neutron scattering. This is the resonant phenomenon in which some certain modes of vibration in the system are amplified by an external driving laser field that satisfies the required resonant frequency conditions and has the intensity approaching some threshold value (see review [10] and references therein). This provides almost a sudden increase in quasiparticle concentrations when the parametric excitation condition is reached, what will strongly influence the values of the scattering cross sections. This effect has been shown theoretically for the process of neutron inelastic scattering on anomalous optical phonons in ref. [6] using an appropriate formalism describing the anomalous fluctuations in electron-phonon systems placed under the action of a high-intensity laser field.

In this work, following the same method, we present the calculations of correlation functions for the polaritons (this means an account of the retardation effect in the Coulomb interaction between particles), in the mentioned non-equilibrium situation, which are used then to obtain the analytical formulae for the cross sections of neutron inelastic scattering on these parametrically excited polaritons. The formulae thus obtained show the resonant behaviour of the cross sections as the intensity of the driving field tends to the threshold value determined by the parametric excitation theory. However, the complicated form of these formulae makes it difficult to have any quantitative representation that is necessary when the question about experimental possibilities is touched.

In a subsequent paper, using the relations obtained in the general parametric excitation theory [10,11], an essential simplification of the theoretical formulae will be made and a quantitative analysis of the process of neutron inelastic scattering by the plasmon and optical phonon modes amplified by a resonant laser field in the crystal of Indium antimonide will be carried out.

It seems to us that such a complete consideration will show clearly how one can use the laser fields in parametric resonant conditions as described above, together with the choice of sufficiently high-flux neutron sources of suitable energies, to overcome the difficulties caused by small quasi-particle concentrations in some concrete inelastic scattering experiments.

2. SCATTERING CROSS SECTIONS

The physical system we have to consider consists of a neutron interacting with an electron-phonon subsystem of a crystalline solid placed under the action of a strong radiation laser field. The Hamiltonian of such a system will be of the form:

$$\mathcal{H}(t) = \epsilon_N + V(\vec{r}) + H(t), \tag{1}$$

where $\epsilon_N = \hat{P}^2/2m_N$ is the Hamiltonian of a neutron with momentum $\vec{P}$, mass m_N and coordinate $\vec{r}$,

$$V(\vec{r}) = \sum_{\ell,s} b_{\ell s}\, \delta(\vec{r} - \vec{R}_{\ell s}) \tag{2}$$

is the neutron-crystal interaction potential ($b_{\ell s}$ is the nuclear scattering amplitude and $\vec{R}_{\ell s}$ the coordinate vector of the atom s in the unit cell ℓ); H(t) is the Hamiltonian of our crystal interacting with a radiation field that, with the retardation interaction taken into account, can be written in the form:

$$H(t) = H_0(t) + H_{int}(t), \tag{2.1}$$

$$H_0(t) = \frac{1}{2m} \sum_{ps} [\vec{p} - \frac{e}{c}\vec{A}_0(t)]^2 a^+_{ps} a_{ps} + \sum_k \omega_k b^+_k b_k + \sum_{q\nu} (qc)\, c^+_{q\nu} c_{q\nu}, \tag{2.2}$$

$$H_{int}(t) = \frac{1}{2} \sum_{\substack{ps \\ p's'q}} \phi_q\, a^+_{p+qs} a^+_{p'-qs'} a_{p's'} a_{ps} - \sum_{\substack{ps \\ p's'q}} \phi_q \langle a^+_{p'-qs} a_{p's'} \rangle a^+_{p+qs} a_{ps} + \sum_{psk} v_k\, a^+_{p+ks} a_{ps} (b_k + b^+_{-k}) - \frac{e}{mc} \sum_{psq} [\vec{p} - \frac{e}{c}\vec{A}_0(t)] \vec{A}(\vec{q}) a^+_{p+qs} a_{ps}. \tag{2.3}$$

Here, $a^+_{ps}(a_{ps})$, $b^+_k(b_k)$, $c^+_{q\nu}(c_{q\nu})$ are the creation (annihilation) operators of an electron of canonical momentum $\vec{p}$, effective mass m, electric charge e and spin s, of a phonon of wave vector $\vec{k}$ and frequency ω_k, and of a photon of wave vector $\vec{q}$ and polarization ν, respectively; $\vec{A}(\vec{k})$ is the spatial Fourier component of the vector potential of internal self-consistent electromagnetic field,

$$\vec{A}(\vec{k}) = \left(\frac{2\pi c}{k}\right)^{1/2} \sum (c_{k\nu} + c^+_{-k\nu}) \vec{\eta}_{k\nu}$$

($\vec{\eta}_{k\nu}$ is the photon polarization vector); $\phi_q = 4\pi e^2/q^2$ and c the light velocity in vacuum; $\vec{A}_0(t)$ is the vector potential of the strong radiation field that in the well-known dipole approximation can be expressed by

$$\vec{A}_0(t) = \frac{c\vec{E}_0}{\omega_0}\cos\omega_0 t \qquad (3)$$

(the corresponding electric field is $\vec{E}_0(t)=\vec{E}_0\sin\omega_0 t$).

In the treatment of the process of neutron scattering by bound nuclei in terms of the space-time correlation functions[12,13] that is the most general one, the differential cross sections for coherent and incoherent inelastic scattering in the Born approximation are given by the formulae:

$$\left(\frac{d^2\sigma}{d\omega\, d\Omega}\right)_{coh} = \frac{B_c^2}{2\pi}\frac{P'}{P}\iint_{-\infty}^{\infty} d\vec{r}\,dt\, e^{i\vec{\kappa}\vec{r}-i\omega t}G_c(\vec{r},t), \qquad (4.1)$$

$$\left(\frac{d^2\sigma}{d\omega\, d\Omega}\right)_{incoh} = \frac{B_i^2}{2\pi}\frac{P'}{P}\iint_{-\infty}^{\infty} d\vec{r}\,dt\, e^{i\vec{\kappa}\vec{r}-i\omega t}G_i(\vec{r},t), \qquad (4.2)$$

where

$$G_c(\vec{r},t) = \frac{1}{(2\pi)^3 N}\sum_{\ell}^{N}\sum_{\ell'}^{N}\int d\vec{\kappa}\, e^{-i\vec{\kappa}\vec{r}}\langle e^{i\vec{\kappa}\vec{R}_\ell(t)}e^{-i\vec{\kappa}\vec{R}_{\ell'}(0)}\rangle, \qquad (4.3)$$

$$G_i(\vec{r},t) = \frac{1}{(2\pi)^3 N}\sum_{\ell}^{N}\int d\vec{\kappa}\, e^{-i\kappa r}\langle e^{i\vec{\kappa}\vec{R}_\ell(t)}e^{-i\vec{\kappa}\vec{R}_\ell(0)}\rangle, \qquad (4.4)$$

and

$$\vec{R}_\ell(t) = S^{-1}(t)R_\ell S(t) \qquad (4.5)$$

with $S(t) = T\exp\{-i\int^{t} dt' H(t')\}$ in the general case of time-dependent Hamiltonians (T is the Dyson chronological operator). In the formulae (4), B_c, B_i are the scattering amplitudes for the coherent and incoherent processes, respectively (for simplicity the Bravais lattice is supposed here, see also formulae (6.5), (6.6) below), $\vec{P}$ and $\vec{P}'$ are the momenta of a neutron in the initial and the final states, respectively, $\omega=(P'^2-P^2)/2m_N$ is the energy transfer (the system of units with $\hbar=1$ is used), N is the number of unit cells in the crystal and the symbol $\langle\ldots\rangle$ means the statistical averaging over states of the solid subsystem.

Introducing the expansion of the atom displacement $\vec{U}_{\ell s}(t)$ from the equilibrium position $\vec{R}^{o}_{\ell s}$ in normal coordinates,

$$\vec{U}_{\ell s}(t) = \vec{R}_{\ell s}(t) - \vec{R}^{o}_{\ell s} = \sum_{qj}(NM\omega_{qj})^{-1/2}\vec{e}_{qj}\, e^{i\vec{q}\vec{R}^{o}_{\ell s}}Q_{qj}(t), \qquad (5)$$

where $\vec{e}_{qj}$ is the polarization vector of the phonon mode j with the wave vector $\vec{q}$ and $Q_{qj} = b_{qj} + b^{+}_{-qj}$ is the phonon coordinate, one can express the cross sections for the one-phonon scattering process in the form:

$$\left(\frac{d^2\sigma}{d\omega\, d\Omega}\right)_{coh} = \frac{N}{8\pi^2}\frac{P'}{P}\sum_j |\Phi_c(\vec{k},\vec{k}',j)|^2 \omega^{-1}_{k'j} \langle Q^2_{k'j}(\omega)\rangle, \qquad (6.1)$$

$$\left(\frac{d^2\sigma}{d\omega\, d\Omega}\right)_{incoh} = \frac{N}{8\pi^2}\frac{P'}{P}\sum_{qj} |\Phi_i(\vec{k},\vec{q},j)|^2 \omega^{-1}_{qj} \langle Q^2_{qj}(\omega)\rangle, \qquad (6.2)$$

where

$$\Phi_c(\vec{k},\vec{k}',j) = \sum_s B_{cs} M_s^{-1/2} (\vec{k}\,\vec{e}_{k'js}) e^{2\pi i \vec{K}\vec{R}^o_s}, \qquad (6.3)$$

$$\Phi_i(\vec{k},\vec{q},j) = \sum_s B_{is} M_s^{-1/2} (\vec{k}\,\vec{e}_{qjs}), \qquad (6.4)$$

$$B_{cs} = \langle b_{\rho_s} \rangle = (\pi\sigma_{cs})^{1/2}, \qquad (6.5)$$

$$B_{is} = b_{\rho_s} - \langle b_{\rho_s} \rangle = (\pi\sigma_{is})^{1/2}. \qquad (6.6)$$

σ_{cs}, σ_{is} are the microscopic nuclear scattering cross sections for coherent and incoherent processes, respectively (see ref. [14]); $\vec{k}' = \vec{k} - 2\pi\vec{K}$ where $\vec{K}$ is the reciprocal lattice wave vector, $\vec{R}^o_s$ the position vector of the atom s in each unit cell.

The influence of the external driving field $\vec{E}_0(t)$ is contained in the phonon correlation functions $\langle Q^2_{qj}(\omega)\rangle$ that must be calculated on the base of the Hamiltonian H(t) (2.1) to (2.3).

3. CORRELATION FUNCTIONS IN THE PARAMETRIC RESONANCE CONDITIONS

In the case of nonequilibrium statistical particle systems described by Hamiltonians dependent on time strongly, as in the case of our electron-phonon-photon system, the correlation functions of fluctuations cannot be calculated by using the fluctuation-dissipation theorem. In the treatment here below, therefore, we shall follow the suitable formalism proposed in ref. [15] that permits one to express in some nontrivial way the correlation functions in this case via the ones for the system of noninteracting particles. We shall not enter into details that have been already described [1,10] but present here the main steps of this calculation procedure.

We first define the fluctuation operator δf_s of the electron distribution as follows:

$$\delta f_s(\vec{p},\vec{p}+\vec{q},t) \equiv a^+_{ps}(t)\,a_{p+qs}(t) - \langle a^+_{ps}\,a_{p+qs}\rangle, \tag{7}$$

where $\langle a^+_{ps}\,a_{p+qs}\rangle = n_0(\vec{p},t)\,\delta_{\vec{q},0}$, $n_0(\vec{p},t)$ is the equilibrium electron distribution function that is time-independent in the collisionless approximation,

$$n_0(\vec{p},t) = n_0(\vec{p}) = \left(\exp\frac{E_p-\mu}{T}+1\right)^{-1} \tag{8}$$

($E_p = p^2/2m$ and μ is the chemical potential of the electrons). As $\langle b_k(t)\rangle = \langle b^+_{-k}(t)\rangle = 0$, the operators $b_k(t)$, $b^+_{-k}(t)$ themselves represent the fluctuations of phonons.

Analogously, we introduce the corresponding operators for the system of noninteracting particles, $b^\circ_k(t)$, $b^{\circ +}_{-k}(t)$ and

$$\delta f^\circ_s(\vec{p},\vec{p}+\vec{q},t) \equiv a^{\circ +}_{ps}(t)\,a^\circ_{p+qs}(t) - n_0(\vec{p})\,\delta_{\vec{q},0}. \tag{9}$$

All time-dependent operators are taken in the Heisenberg representation, that is,

$$a_{ps}(t) = S^{-1}(t)\,a_{ps}\,S(t), \quad b_k(t) = S^{-1}(t)\,b_k\,S(t), \tag{10.1}$$

$$a^\circ_{ps}(t) = S^{\circ -1}(t)\,a_{ps}\,S^\circ(t), \quad b^\circ_k(t) = S^{\circ -1}(t)\,b_k\,S^\circ(t), \tag{10.2}$$

where

$$S(t) \equiv \exp\{-i\int_{-\infty}^{t} dt'\,H(t')\}, \quad S^\circ(t) \equiv \exp\{-i\int_{-\infty}^{t} dt'\,H_0(t')\}. \tag{10.3}$$

The equations of motion for the just introduced operators are easily obtained on the base of the Hamiltonian (2.1) to (2.3) by using the density-matrix formalism according to which one has at least

$$-i\frac{\partial}{\partial t}\hat{O}(t) = S^{-1}(t)[H(t),\hat{O}]\,S(t) \tag{11}$$

for an arbitrary physical operator $\hat{O}(t)$. The equations of motion for the fluctuation operators of noninteracting particles lead directly to the following expressions for the correlation functions of the quantities δf°_s, $b^\circ_k(t)$ and $b^{\circ +}_{-k}(t)$:

$$\langle \delta f^{\circ +}_{s'}(\vec{p}',\vec{p}'+\vec{q}',t')\,\delta f^\circ_s(\vec{p},\vec{p}+\vec{q},t)\rangle = \delta_{\vec{p}\vec{p}'}\,\delta_{\vec{q}\vec{q}'}\,\delta_{ss'} \times \tag{12.1}$$

$$n_0(\vec{p}+\vec{q})\,[1-n_0(\vec{p})]\exp\{i\lambda(\sin\omega_0 t - \sin\omega_0 t') + i(E_p - E_{p+q})(t-t')\},$$

$$\langle Q^{o+}_{k'}(t')Q^{o}_{k}(t)\rangle = \delta_{\vec{k}\vec{k}'}\,[n_k e^{-i\omega_k(t-t')} + (n_k+1)e^{i\omega_k(t-t')}], \qquad (12.2)$$

where

$$Q^{o}_{k}(t) = b^{o}_{k}(t) + b^{o+}_{-k}(t),\quad n_k = \langle b^{+}_{k} b_{k}\rangle = (e^{\omega_k/T}-1)^{-1}, \qquad (12.3)$$

and

$$\lambda = e(\vec{q}\cdot\vec{E}_0)/m\omega_0^2 .$$

To establish the relations between the correlation functions (12) and the ones of the system of interacting particles in the presence of the high-power laser field the following quantities are to be introduced:

$$\tilde{f}(\vec{p},\vec{p}+\vec{q},t) \equiv a^{+}_{ps}(t)\,a_{p+qs}(t) - a^{o+}_{ps}(t)\,a^{o}_{p+qs}(t), \qquad (13.1)$$

$$\tilde{b}_k(t) \equiv b_k(t) - b^{o}_{k}(t), \qquad (13.2)$$

$$\tilde{b}^{+}_{-k}(t) = b^{+}_{-k}(t) - b^{o+}_{-k}(t), \qquad (13.3)$$

the equations of motion for which can be written in the form:

$$-i\frac{\partial}{\partial t}\tilde{f}_s(\vec{p},\vec{p}+\vec{q},t) = (E_p - E_{p+q} + \omega_0\lambda\cos\omega_0 t)\,\tilde{f}_s(\vec{p},\vec{p}+\vec{q},t) +$$

$$+[n_0(\vec{p}+\vec{q}) - n_0(\vec{p})]\{\phi_q \sum_{p's'}\delta f_{s'}(\vec{p}',\vec{p}'+\vec{q},t) - \qquad (14.1)$$

$$-\frac{e}{mc}[\vec{p} - \frac{e}{c}\vec{A}_0(t)]\vec{A}(\vec{q},t) + v_q[Q^{o}_{q}(t) + \tilde{b}_q(t) + \tilde{b}^{+}_{-q}(t)]\},$$

$$-i\frac{\partial}{\partial t}\tilde{b}_k(t) = -\omega_k \tilde{b}_k(t) - v_{-k}\sum_{p's'}\delta f_{s'}(\vec{p}',\vec{p}'+\vec{k},t), \qquad (14.2)$$

$$-i\frac{\partial}{\partial t}\tilde{b}^{+}_{-k}(t) = \omega_k \tilde{b}^{+}_{-k}(t) - v_{-k}\sum_{p's'}\delta f_{s'}(\vec{p}',\vec{p}+\vec{k},t), \qquad (14.3)$$

with the initial conditions:

$$\lim_{t\to-\infty}\tilde{f}(t) = \lim_{t\to-\infty}\tilde{b}_k(t) = \lim_{t\to-\infty}\tilde{b}^{+}_{-k}(t) = 0. \qquad (15)$$

A rigorous analysis of this system of equations leads to the following equation for the density fluctuation operator $\delta n(\vec{q},t) = \sum_{ps}\delta f_s(\vec{p},\vec{p}+\vec{q},t)$:

$$\delta n(\vec{q},t) - i\int_{-\infty}^{t} dt'\,\Omega_q(t,t')\{\phi_q\delta n(\vec{q},t') - \frac{e}{mc}[\vec{p} - \frac{e}{c}\vec{A}_0(t')]\vec{A}(\vec{q},t') +$$

$$|v_q|^2\int_{-\infty}^{t'} dt''\,\Gamma_q(t'-t'')\delta n(\vec{q},t'')\} = \delta n^{o}(\vec{q},t) + i\int_{-\infty}^{t} dt'\,\Omega_q(t,t')v_q\,Q^{o}_{q}(t'), \qquad (16)$$

with the notation:

$$\Omega_q(t,t') \equiv e^{i\lambda(\sin\omega_0 t - \sin\omega_0 t')} \sum_{ps} [n_0(\vec{p}+\vec{q}) - n_0(\vec{p})] e^{i(E_p - E_{p+q})(t-t')}, \quad (17.1)$$

$$\Gamma_q(t'-t'') \equiv i\{ e^{i\omega_q(t'-t'')} - e^{-i\omega_q(t'-t'')} \}, \quad (17.2)$$

$$\delta n^\circ(\vec{q},t) \equiv \sum_{ps} \delta f_s^\circ (\vec{p},\vec{p}+\vec{q},t). \quad (17.3)$$

Now, to have the closed system of equations to solve for the correlation functions of physical operators, one must include into consideration the system of Maxwell equations for the self-consistent electromagnetic field with the vector potential $\vec{A}(\vec{q},t)$ and the scalar potential $\phi(\vec{q},t)$ expressed by

$$\phi(\vec{q},t) = \phi_q \delta n(\vec{q},t) + v_q Q_q(t). \quad (18)$$

We present this system of equations in the form:

$$\Delta\vec{A} - \frac{1}{c}\hat{\epsilon}\frac{\partial}{\partial t}\left(\frac{1}{c}\frac{\partial\vec{A}}{\partial t} + \vec{\nabla}\phi\right) = -\frac{4\pi}{c}\vec{j}, \quad (19.1)$$

$$\operatorname{div}\hat{\epsilon}\left(\frac{1}{c}\frac{\partial\vec{A}}{\partial t} - \vec{\nabla}\phi\right) = -4\pi\rho, \quad (19.2)$$

$$\operatorname{div}\vec{A} = 0, \quad (19.3)$$

where

$$\vec{j}(\vec{k},t) = \frac{e}{m}\sum_{ps}\left[\vec{p} + \frac{\vec{k}}{2} - \frac{e}{c}\vec{A}_0(t)\right]\delta f_s(\vec{p},\vec{p}+\vec{k},t) - \frac{e^2}{mc}\sum_{ps}\vec{A}(\vec{k},t)n_0(\vec{p}), \quad (19.4)$$

$$\rho(\vec{k},t) = e\sum_{ps}\delta f_s(\vec{p},\vec{p}+\vec{k},t). \quad (19.5)$$

Here, $\hat{\epsilon}$ is the dielectric tensor of the crystal lattice. Further, from the equations of motion for $\tilde{f}$ the fluctuation operator δf_s that enters into equations (19) can be expressed via δf_s° by:

$$\delta f_s(\vec{p},\vec{p}+\vec{q},t) = \delta f_s^\circ(\vec{p},\vec{p}+\vec{q},t) + e^{i[(E_p - E_{p+q})t + \lambda\sin\omega_0 t]} \times$$

$$-\int_{-\infty}^{t} dt'[n_0(\vec{p}+\vec{q})-n_0(\vec{p})]\{\phi_q \delta n(\vec{q},t') - \frac{e}{mc}[\vec{p}-\frac{e}{c}\vec{A}_0(t')]\vec{A}(\vec{q},t') - v_q Q^{\circ}_q(t') + |v_q|^2 \int_{-\infty}^{t'} dt'' \Gamma_q(t'-t'')\delta n(\vec{q},t'')\} e^{-i[(E_p - E_{p+q})t' + \lambda \sin\omega_0 t']} \tag{20}$$

In the presence of the retardation effect, as has been shown by the parametric excitation theory [10,11], the coupling between longitudinal and transverse waves of the electron-phonon system takes place in the geometry with $\vec{E}_0 \perp \vec{k}$ where $\vec{k}$ is the wave vector of the modes considered. Taking this case into consideration and passing to the Fourier representation of physical quantities, we obtain from the systems of equations (16), (19), (20) the equations for the Fourier components of δn and $\vec{A}$ as follows:

$$\delta n(\vec{q},\omega)\mathcal{E}_\ell(\vec{q},\omega) - e\lambda' P_q(\omega)[A(\omega+\omega_0)+A(\omega-\omega_0)] = \delta n^{\circ}(\vec{q},\omega) + v_q P_q(\omega) Q^{\circ}_q(\omega), \tag{21.1}$$

$$A(\vec{q},\omega)\left[\frac{q^2c^2}{\omega^2} - \mathcal{E}_t(\vec{q},\omega)\right] = \frac{4\pi c}{\omega^2} j_0(\vec{q},\omega) - \frac{4\pi e c^2}{\omega^2}\lambda'\{\delta n(\vec{q},\omega+\omega_0) + \delta n(\vec{q},\omega-\omega_0)\}, \tag{21.2}$$

where $A(\vec{q},\omega)$ and $j_0(\vec{q},\omega)$ here mean the components of the vectors $\vec{A}$ and $\vec{j} = (e/m)\sum_{ps} \vec{p}\,\delta f^{\circ}_s(\vec{p},\vec{p}+\vec{q},t)$ parallel to the external electric field vector $\vec{E}_0$, and the following notation has been used:

$$\mathcal{E}_\ell(\vec{q},\omega) = 1 - \phi_q P_q(\omega) - |v_q|^2 P_q(\omega) D_q(\omega), \tag{22.1}$$

$$\mathcal{E}_t(\vec{q},\omega) = \epsilon(\vec{q},\omega) - \frac{\omega_p^2}{\omega^2} + i\frac{4\pi e^2 c^2}{\omega^2} R_q(\omega), \tag{22.2}$$

$$P_q(\omega) = \sum_{ps} \frac{n_0(\vec{p}+\vec{q}) - n_0(\vec{p})}{E_{p+q} - E_p - \omega - i0}, \tag{22.3}$$

$$R_q(\omega) = \frac{1}{m^2c^2} \sum_{ps} \frac{[n_0(\vec{p}+\vec{q}) - n_0(\vec{p})]p_\perp^2}{E_{p+q} - E_p - \omega - i0}, \tag{22.4}$$

$$D_q(\omega) = \frac{1}{\omega - \omega_q + i0} - \frac{1}{\omega + \omega_q + i0}, \quad (22.5)$$

$$\lambda' = \frac{eE_0}{2mc\omega_0}. \quad (22.6)$$

We now apply the following resonant condition concerning the frequencies:

$$\omega_0 = \omega_\ell + \omega_t, \quad (23)$$

where ω_ℓ and ω_t are determined from the dispersion equations $\mathrm{Re}\,\mathcal{E}_\ell(\vec{q},\omega)=0$ and $\mathrm{Re}\Delta(\vec{q},\omega) = \mathrm{Re}[q^2c^2/\omega^2 - \mathcal{E}_t(\vec{q},\omega)] = 0$, respectively, and represent the frequencies of longitudinal (ω_ℓ) and transverse (ω_t) modes of the electron-phonon system for the fixed value of the wave vector $\vec{k}$. Thus, we have

$$\omega^2_{\ell\,1,2} = \frac{1}{2}\{\omega_p^2 + \omega_{L0}^2 \pm (\omega_p^2 - \omega_{L0}^2)[1 + \frac{8\phi_q^{-1}|v_q|^2\omega_{L0}\omega_p^2}{(\omega_p^2-\omega_{L0}^2)^2}]^{1/2}\}, \quad (24)$$

and $\omega^2_{t\,1,2}$ is of the same form (24) with $\omega^2_{pc} = (\omega_p^2 + q^2c^2)/\epsilon_\infty$ instead of the quadratic plasma frequency $\omega_p^2 = 4\pi e^2 n/\epsilon_\infty m$ in (24) (ϵ_∞ is the high-frequency dielectric constant of the crystal lattice and ω_{L0} the longitudinal optical phonon frequency).

The condition (23) expresses the so-called two-mode approximation in which the equations can be considerably simplified. The calculations in this case lead to the following formula for the correlation function of electron density fluctuations:

$$\langle\delta n^+(\vec{q}',\omega')\delta n(\vec{q},\omega)\rangle = 2\pi\delta_{\vec{q}\vec{q}'}\delta_{\omega\omega'}\langle\delta n^2(\vec{q},\omega)\rangle, \quad (25.1)$$

$$\langle\delta n^2(\vec{q},\omega)\rangle = \theta(\vec{q},\omega)\ \{|F_1(\vec{q},\omega,T,\lambda') -$$

$$F_2(\vec{q},\omega,T,\lambda')(\omega_0 - |\omega|)\sum_i[\delta(|\omega| - \omega_0 + \omega_{ti}) + \delta(|\omega| - \omega_0 - \omega_{ti})]|\}\times \quad (25.2)$$

$$\sum_i[\delta(\omega + \omega_{\ell_i}) + \delta(\omega - \omega_{\ell_i})],$$

where the notation is as follows:

$$\theta(\vec{q},\omega) \equiv \pi\left|\frac{\omega(\omega^2-\omega_{L0}^2)}{\omega_{\ell_1}^2 - \omega_{\ell_2}^2}\right|\frac{|v_q|^2P_q^2(|\omega|)\,\mathrm{Im}\,D_q(|\omega|) - \mathrm{Im}\,P_q(|\omega|)}{\mathrm{Im}\ \mathcal{E}_\ell(\vec{q},|\omega|)}, \quad (26.1)$$

$$F_1(\vec{q},\omega,T,\lambda')=|1-I^2(\vec{q},\omega,\lambda')|^{-1}(e^{\omega/T}-1)^{-1}, \tag{26.2}$$

$$F_2(\vec{q},\omega,T,\lambda')=|1-I^2|^{-1}\tilde{I}^2(\vec{q},\omega,\lambda')(e^{(\omega-\omega_0 \mathrm{sgn}\omega)/T}-1)^{-1}, \tag{26.3}$$

$$I^2(\vec{q},\omega,\lambda') \equiv \frac{8\pi\omega_p^2(ec\lambda')^2}{\phi_q\omega^2(\omega-\omega_0)^2 \,\mathrm{Im}\,\mathcal{E}_\ell(\vec{q},|\omega|)\,\mathrm{Im}\,\Delta(\vec{q},\omega_0-|\omega|)}, \tag{26.4}$$

$$\tilde{I}^2(\vec{q},\omega,\lambda') \equiv I^2\frac{\pi}{4}\frac{\omega^2}{\omega_p^2}\left|\frac{(|\omega|-\omega_0)^2-\omega_{T0}^2}{\epsilon_\infty(\omega_{t1}^2-\omega_{t2}^2)}\right| \times$$
$$\frac{\phi_q P_q^2(|\omega|)[\epsilon(\vec{q},\omega_0-|\omega|)-\omega_{pc}^2/(\omega_0-|\omega|)^2]\,\mathrm{Im}\,\mathcal{E}_\ell(\vec{q},|\omega|)}{|v_q|^2P_q^2(|\omega|)\,\mathrm{Im}\,D_q(|\omega|)-\mathrm{Im}\,P_q(|\omega|)}. \tag{26.5}$$

Note that $i=1,2$ and ω_{T0}in (26.4) is the transverse optical phonon frequency.

The correlation function for phonon fluctuations is now obtained without difficulties by using the relation:

$$Q_q(\omega)=Q^\circ_q(\omega)+v_{-q}D_q(\omega)\delta n(\vec{q},\omega), \tag{27}$$

and is of the form:

$$\langle Q^+_{q'}(\omega')Q_q(\omega)\rangle = 2\pi\delta_{\vec{q}\vec{q}'}\delta_{\omega\omega'}\langle Q_q^2(\omega)\rangle, \tag{28.1}$$

$$\langle Q_q^2(\omega)\rangle=\theta_Q(\vec{q},\omega,\lambda')\{|F_{1Q}(\vec{q},\omega,T,\lambda')-$$
$$F_{2Q}(q,\omega,T,\lambda')(\omega_0-|\omega|)\sum_i[\delta(|\omega|-\omega_0+\omega_{ti})+\delta(|\omega|-\omega_0-\omega_{ti})]|\}\times \tag{28.2}$$
$$\sum_i[\delta(\omega+\omega_{\ell i})+\delta(\omega-\omega_{\ell i})]$$

with the following notation used:

$$\theta_Q(\vec{q},\omega,\lambda')\equiv\pi\left|\frac{\omega(\omega^2-\omega_{L0}^2)}{\omega_{\ell 1}^2-\omega_{\ell 2}^2}\right||\mathrm{Im}\,\mathcal{E}_\ell(\vec{q},|\omega|)|^{-1}\times$$
$$\{[1-I^2(\lambda')+2\mathrm{Re}(1-\phi_qP_q(|\omega|))]\mathrm{Im}\,D_q(|\omega|)+ \tag{29.1}$$
$$|v_q|^2|D_q(|\omega|)|^2[|v_q|^2P_q^2(|\omega|)\mathrm{Im}D_q(|\omega|)\tilde{\theta}(\vec{q},\omega,\lambda')-\mathrm{Im}\,P_q(|\omega|)]\},$$

$$F_{2Q}(\vec{q},\omega,T,\lambda')=|1-I^2(\vec{q},\omega,\lambda')|^{-1}\tilde{I}^2_Q(\vec{q},\omega,\lambda')(e^{(\omega-\omega_0\mathrm{sgn}\omega)/T}-1)^{-1}, \quad (29.2)$$

$$\tilde{I}^2_Q(\vec{q},\omega,\lambda')=I^2(\vec{q},\omega,\lambda')\frac{\pi}{4}\frac{\omega^2}{\omega_p^2}\left|\frac{(|\omega|-\omega_0)^2-\omega^2_{T0}}{\epsilon_\infty(\omega^2_{t1}-\omega^2_{t2})}\right|\times$$

$$\times\frac{\phi_q P_q^2(|\omega|)[\epsilon(\vec{q},(\omega_0-|\omega|))-\omega^2_{pc}/(\omega_0-|\omega|)^2]\mathrm{Im}\,\mathcal{E}_\ell(\vec{q},|\omega|)}{|v_q|^{-2}|D_q(|\omega|)|^{-2}\mathrm{Im}D_q(|\omega|)[1-I^2+2\mathrm{Re}(1-\phi_qP_q(|\omega|))]+|v_q|^2P_q^2(|\omega|)\mathrm{Im}D_q(|\omega|)\tilde{\theta}-\mathrm{Im}P_q(|\omega|)}, \quad (29.3)$$

$$\tilde{\theta}(\vec{q},\omega,\lambda')=1-I^2(q,\omega,\lambda')\frac{\phi_q}{|v_q|^2}\frac{\omega^2(\omega^2-\omega^2_{L0})}{\omega_{L0}\omega_p^2}, \quad (29.4)$$

where $F_{1Q}(\vec{q},\omega,T,\lambda')=F_1(\vec{q},\omega,T,\lambda')$ and $I^2(\vec{q},\omega,\lambda')$ is defined by (26.4).

From the formulae (25) and (28) we see that the spectral distributions of fluctuations reveal the sharp maxima at the frequencies of the coupled longitudinal plasmon-phonon as well as the coupled transverse polariton modes of the system which could be excited in pairs (one longitudinal and one transverse modes each time) by the laser field. Besides that, the obtained formulae show that the level of collective fluctuations increases anomalously when approaching the border line of parametric instability regions for which the term $1-I^2\to 0$. By the definitions (6) the same features will characterize the behaviour of the cross sections of neutron inelastic scattering by the plasmon-phonon and polariton modes in the crystal.

In the case of $\vec{k}\parallel\vec{E}_0$ geometry the transverse and longitudinal modes of the system do not couple to each other, and only the longitudinal modes represent the practical interest in the problem considered since the pure transverse vibrations are rather "hard" to be excited by radiation fields (see refs.[10,11]). In this case the spectral distribution of the phonon correlation function that exhibits the main features of the neutron inelastic scattering process by the coupled longitudinal plasmon-phonon modes will have the form (28) with the following notation (the same formula has been obtained in ref.[6]):

$$\theta_Q \to \theta_\ell(\vec{q},\omega,\lambda) \equiv 2\pi \left| \frac{\omega_{L0}(\omega^2-\omega_p^2)}{\omega(\omega_{\ell 1}^2-\omega_{\ell 2}^2)} \right| \times$$

$$\left[1-\frac{\alpha(\omega)}{\alpha(\omega_0-|\omega|)} I_\ell^2(\vec{q},\omega,\lambda) \frac{\mathrm{Im}\,\Pi_q(|\omega|)}{\mathrm{Im}\,\Pi_q(\omega_0-|\omega|)}\right]^{-1}, \tag{30.1}$$

$$\alpha(\omega) \equiv \frac{\omega^2-\omega_p^2}{(\omega_{\ell 1}+|\omega|)(\omega_{\ell 2}+|\omega|)}, \quad \Pi_q(\omega) \equiv \frac{\mathcal{E}_\ell(\vec{q},\omega)}{[1-\phi_q P_q(\omega)]D_q(\omega)}, \tag{30.2}$$

$$I_\ell^2(\vec{q},\omega,\lambda) \equiv J_1^2(\lambda)|v_q|^2[1-\phi_q P_q(|\omega|)][1-\phi_q P_q(\omega_0-|\omega|)]\times$$

$$\frac{[P_q(|\omega|)-P_q(\omega_0-|\omega|)][D_q^{-1}(|\omega|)-D_q^{-1}(\omega_0-|\omega|)]}{\mathrm{Im}\,\Pi_q(|\omega|)\,\mathrm{Im}\,\Pi_q(\omega_0-|\omega|)}, \tag{30.3}$$

where $J_1(\lambda)$ is the Bessel function of the first kind of the argument $\lambda \equiv e(\vec{q}\vec{E}_0)/m\omega_0^2$; the term with F_{1Q} and F_{2Q} in braces in (28.2) should be replaced by F_ℓ,

$$F_\ell(\vec{q},\omega,T,\lambda) = |1-I^2(\vec{q},\omega,\lambda)|^{-1}\{|(e^{\omega/T}-1)^{-1} - I^2(\vec{q},\omega,\lambda)(e^{(\omega-\omega_0 \mathrm{sgn}\omega)/T}-1)^{-1}|\} . \tag{30.4}$$

In a subsequent paper this last case, as the most simple one, will be analysed to obtain some numerical estimations of the scattering cross sections on plasmon-phonon modes in a crystal of indium antimonide.

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