Neutron scattering in molecular crystals at a high level of optical pumping of excitons

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Neutron scattering in molecular crystals in the presence of a high exciton density is theoretically considered. Two possible channels of scattering by excitons are investigated: magnetic scattering by triplet excitons, and nuclear scattering. It is shown that the change of the nuclear configurations of molecules excited by electrons leads, as a result of the correlation of the excitations, to the appearance of an additional channel of nuclear inelastic scattering. The scattering of neutrons by an excition system of high density is considered with allowance for the dynamic and kinematic exciton-exciton interactions. The features of inelastic scattering of neutrons in the presence of a Bose condensate of molecular excitons is also considered. It is shown that magnetic scattering of high-energy neutrons can be used to detect an exciton condensate. In view of the discrepancy between the results of Agranovich and Lalov [Sov. Phys. JETP **42**, 328 (1975)], Krivoglaz and Rashba [Sov. Phys. Solid State **21**, 1705 (1979)], and Elliott and Shukla [J. Phys. C12, 5463 (1979)], an investigation is made of the production, due to electron-phonon interaction, of excitons in inelastic neurton scattering. The formulas obtained agree with the results of Krivoglaz and Rashba.

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1. INTRODUCTION

Inelastic scattering of slow neutrons is traditionally used as a source of information on the energy spectrum of elementary excitations of condensed systems. Neutron scattering by various quasiparticles is the subject of a large number of papers, both theoretical and experimental (see, e.g., Refs. 1–3).

Neutron scattering by excitons was first considered by Glauberman and one of us in Ref. 4, where we studied the principal mechanisms of the interaction of thermal neutrons with Wannier-Mott excitons in semiconductors. Neutron scattering with exciton production in molecular crystals was investigated in Refs. 5–7; an operator formalism was used in Ref. 8 to take into account the various channels of neutron scattering by a one-dimensional molecular chain. Krivoglas and Rashba[6] have recently demonstrated the feasibility in principle of determining directly the exciton dispersion law from the inelastic neutron-scattering spectra.

Scattering by impurity centers of crystals, besides the direct excitation of electronic states by neutrons via the direct magnetic mechanism, involves also an indirect excitation mechanism due to the electron-vibrational interaction that mixes the electronic and vibrational states.[9] An analog of such a mechanism in the case of delocalized (extended) excitations is exciton production in neutron scattering, as a result of the exciton-phonon interaction considered in Refs. 5–7. It must be noted, however, that although the mechanism responsible for the exciton production is the same in the cited papers, the analytic expressions obtained in these references for the scattering cross sections are different. The ratio of the cross section for scattering with exciton production to the cross section with phonon production is estimated by Agranovich and Lalov[5] at $4\hbar\omega_k\gamma_k^2/\varepsilon_k^2$, from the results of Krivoglaz and Rashba[6] it follows that this ratio is $4\hbar^2\omega_k^2\gamma_k^2/\varepsilon_k^4$, and according to Ref. 7 it amounts to $\gamma_k^2/\varepsilon_k^2$ (ω_k is the phonon frequency, ε_k is the exciton energy, and γ_k are the exciton-phonon interaction constants); the reasons for the differences are not discussed by the authors. An analysis made in this connection in the present paper (see the last section) agrees with the results of Krivoglaz and Rashba.

Modern laser-pumping methods provide sufficiently high densities of excited molecules in a crystal, $\sim 5\times 10^{-4}$ of the total number of molecules.[10–12] In the present study we have investigated the mechanisms of neutron scattering in molecular crystals with a high density of excitons produced by a high-power laser pulse. Two possible channels of scattering by the excitons are considered: magnetic scattering by triplet excitons, and nuclear scattering that takes place for triplet as well as singlet excitons. When neutron scattering by nuclei is considered, account must be taken of the change of the equilibrium positions of the nuclei following the electronic excitation of the molecules. The last circumstance, owing to the correlations between the electronic excitations, leads to the appearance of an additional nuclear-scattering channel with a change of the exciton state. According to the estimates presented below for transfer energies $E\sim 0.1$ eV, this yields $0.1\bar{n}$ of the scattering cross section in the absence of excitation ($\bar{n}$ is the average exciton density).

In view of the possible Bose condensation of excitons,[13–16] we have considered neutron scattering in the presence of a condensate of molecular excitons. The distinguishing features of neutron scattering under Bose-condensation conditions are closely connected with the interaction of the excitons and their statistics. The main type of transition caused by neutrons is one with production of above-condensate elementary excitations, which makes it possible in principle to determine directly their energy spectrum from the inelastic-scattering data. The cross section for magnetic scattering of neutrons with production and absorption of excitons in

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a Bose condensate contains two δ-like peaks of equal intensity. Observation of these maxima in experiment would confirm the formation of the condensate.

2. NEUTRON SCATTERING BY EXCITONS. CASE OF ABSENCE OF EXCITON PHONON INTERACTION

We describe the exciton subsystem in the two-level approximation by the Hamiltonian

$$H_{ex}=\sum_k(\varepsilon_k-\mu)a_k^+a_k+\sum_k F_k\rho_k\rho_{-k}, \qquad (1)$$

where $\varepsilon_k=\Delta+Q_k$, Δ is the excitation energy of the molecule in the crystal, Q_k is expressed in terms of the matrix elements of the molecule interaction potential, μ is the chemical potential of the excitons,

$$\rho_k=\mathfrak{N}^{-1/2}\sum_p a_p^+a_{p+k} \qquad (2)$$

is the collective exciton-density operator, and $\mathfrak{N}$ is the number of sites in the crystal. We confine ourselves for simplicity to crystals with one molecule per unit cell. The term with F_k in (1) describes the dynamic interaction of the exciton. The kinematic interaction will be taken into account with the aid of exact commutation relations for the exciton operators.[18] It is convenient to represent them in the form[16,19]

$$\begin{aligned}&[a_k,a_p^+]=\delta_{kp}-2\mathfrak{N}^{-1/2}\rho_{k-p},\\&[a_k,\rho_p]=\mathfrak{N}^{-1/2}a_{k+p},\\&[a_k^+,\rho_p]=-\mathfrak{N}^{-1/2}a_{k-p}^+.\end{aligned} \qquad (3)$$

The dynamic and kinematic exciton-exciton interactions become significant at high exciton densities.

In this section we consider the scattering of neutrons of energy ≲ 1 eV by excitons. We shall call scattering by excitons the channel connected with the difference between the amplitudes of scattering by the crystal molecules in the excited and ground electronic states. The principal mechanisms of scattering by excitons are nuclear scattering, due to the change of the equilibrium nuclear configurations in electronic excitation of the molecules, and magnetic scattering, which is determined by the difference between the magnetic interaction (spin-spin or spin-orbit) of the neutron with the molecules in the excited and ground states. The latter mechanism is particularly important for molecules excited into a state with a total electron spin $S\neq 0$ (for example, triplet excitons). The probability of electronic transitions as a result of magnetic interaction of a neutron with electrons in the indicated neutron-energy region is much less that 10^{-6} of the nuclear scattering, because of the smallness of the magnetic form factor of the inelastic scattering.[6,7] If the exciton-phonon interaction is disregarded, the number of excitons in the scattering process can be regarded as constant.

The potential of the interaction with the nuclei is chosen in the Born approximation in the form of the Fermi quasipotential[1]

$$V_{nuc}(\mathbf{r}_n)=\sum_{\nu j}C_{\nu j}\delta(\mathbf{r}_n-\mathbf{R}_{\nu j}), \qquad (4)$$

where $\mathbf{r}_n$ and $\mathbf{R}_{\nu j}$ are the radius vector of the neutron and of the nucleus j located in the ν-th unit cell,

$$C_{\nu j}=A_{\nu j}+2[s_{\nu j}(s_{\nu j}+1)]^{-1/2}B_{\nu j}(\mathbf{s}_n\mathbf{s}_{\nu j}), \qquad (5)$$

$A_{\nu j}$ and $B_{\nu j}$ are constants that describe the interaction of the neutron with the nucleus,[20] $\mathbf{s}_n$ and $\mathbf{s}_{\nu j}$ are the spin operators of the neutron and of the nucleus. We represent the radius vector of the nucleus by the sum $\mathbf{R}_{\nu j}=\mathbf{R}_{0,\nu j}+\mathbf{u}_{\nu j}$, where $\mathbf{R}_{0,\nu j}=\mathbf{R}_{0,\nu}+\mathbf{R}_{0,j}$ while $\mathbf{R}_{0,\nu}$ is the equilibrium coordinate of the mass center of the molecule, $\mathbf{R}_{0,j}$ is the equilibrium coordinate of the nucleus reckoned from the mass center, and $\mathbf{u}_{\nu,j}$ is the shift of the nucleus from the equilibrium position. We separate in the vector $\mathbf{u}_{\nu,j}$ the displacement $\mathbf{u}_\nu$ as the molecule as a whole: $\mathbf{u}_{\nu j}-\mathbf{u}_\nu+\mathbf{w}_{\nu j}$.

The matrix element of the potential of the interaction of the neutron with the molecules in terms of the neutron wave functions is represented in the form

$$V_{nuc}(\mathbf{q})=\sum_\nu a_\nu(\mathbf{q})\exp(iqR_\nu), \qquad (6)$$

where $\mathbf{R}_\nu=\mathbf{R}_{0,\nu}+\mathbf{u}_\nu$, $\mathbf{q}=\mathbf{p}_1-\mathbf{p}_2$ is the momentum transfer to the neutron (we use a system of units with $\hbar=1$),

$$a_\nu(\mathbf{q})=\sum_j C_{\nu j}\exp[i\mathbf{q}(\mathbf{R}_{0,j}+\mathbf{w}_{\nu j})]. \qquad (7)$$

We shall assume that the coordinates $\mathbf{R}_{0,j}$ in (7) pertain the ground state of the molecule. In the general case, as a result of excitation of the molecule, the equilibrium coordinates of the nuclei acquire an increment $\Delta\mathbf{R}_{0,j}=\mathbf{R}_{0,}^{\ j}-\mathbf{R}_{0,j}$. The corresponding change of the quantity a_ν ($\mathbf{q}$) (7) (provided that we neglect the dependence of the displacement $\mathbf{w}_{\nu j}$ on the electronic state of the molecule) is

$$\Delta a_\nu(\mathbf{q})\approx i\mathbf{q}\sum_j\Delta\mathbf{R}_{0,j}A_{\nu j}\exp[i\mathbf{q}(\mathbf{R}_{0,j}+\mathbf{w}_{\nu j})]. \qquad (8)$$

As a result there is added to the matrix element $V_{nuc}(\mathbf{q})$ (6) a term connected with the change of a_ν in the excited state. It can be written down by introducing the number of excitations on a site $N_\nu=b_\nu^+b_\nu$, where b_ν^+ and b_ν are the operators of creation and annihilation of excitations in the site representation. In place of (6) we obtain

$$V_{nuc}(\mathbf{q})=\sum_\nu(a_\nu(\mathbf{q})+\Delta a_\nu(\mathbf{q})N_\nu)\exp(i\mathbf{q}\mathbf{R}_\nu). \qquad (9)$$

The first term in (9) determines the scattering in the absence of electronic excitations in the crystal. The second is connected with the corrections to the interaction potential because of the presence of excited molecules and corresponds to the nuclear channel of scattering by excitons.

The matrix element of the spin-spin interaction of a neutron with excited molecules in a state with $S\neq 0$ is expressed in terms of the magnetic form factor of the molecule $\chi_\nu(\mathbf{q})$ (Ref. 3) and is equal to

$$V_M(q)=-\frac{4\pi r_e\gamma_n}{m_n}\sum_\nu\chi_\nu(\mathbf{q})\,(\mathbf{S}_\nu\mathbf{I}_n)N_\nu\exp(i\mathbf{q}\mathbf{R}_\nu), \qquad (10)$$

where m_n is the neutron mass, r_e is the electromagnetic radius of the electron, $\gamma_n=-1.913$ is the magnetic moment of the neutron in nuclear magnetons, $\mathbf{S}_\nu$ is the operator of the total electron spin of the molecule, $\mathbf{I}_n=\mathbf{s}_n-\mathbf{e}(\mathbf{s}_n\cdot\mathbf{e})$, and $\mathbf{e}=\mathbf{q}/q$. The effective process for a noticeable contribution of the magnetic scattering by triplet electrons to the total scattering cross section in the crystal should be the singlet-triplet conversion process.

We express the formulas for the scattering by excitons in terms of the cross section for coherent and incoherent scattering by individual nuclei with allowance for the averaging over the isotopes[20]

$$\sigma_j^c=m_n^2\pi^{-1}|\bar{A}_j|^2,\quad \sigma_j^{inc}=m_n^2\pi^{-1}(\overline{|A_j-\bar{A}_j|^2}+\overline{B_j^2})$$

and in terms of the quantities

$$\tau_j^c=m_n^2\pi^{-1}(\mathbf{q}\Delta\mathbf{R}_{0,j})^2|\bar{A}_j|^2,\quad \tau_j^{inc}=m_n^2\pi^{-1}(\mathbf{q}\Delta\mathbf{R}_{0,j})^2\overline{|A_j-\bar{A}_j|^2},$$

which are connected with the deformation of the nuclear core upon excitation. We introduce also the cross section for magnetic scattering by a molecule with $S=1$, calculated per unit solid angle

$$\sigma_\nu^M(\mathbf{q})=4r_e^2\gamma_n^2\overline{\chi_\nu^2(q)}/3.$$

The cross sections for the scattering of neutrons by excitons are best calculated on the basis of the time-dependent Van Hove formalism.[3,19] In the course of the calculations we shall assume that the neutron beam is not polarized, therefore the nuclear and magnetic scatterings do not interfere. The general formula for the cross section for scattering of neutrons with interaction potentials (9) and (19) is of the form

$$\frac{\delta^2\Sigma}{\delta E\delta\Omega}=\frac{1}{16\pi^2}\frac{p_2}{p_1}\int_{-\infty}^{\infty}dt\,e^{-iEt}\Big\{\sum_{\nu\nu'}\sum_{jj'}\exp[-i\mathbf{q}(\mathbf{R}_{0,\nu j}-\mathbf{R}_{0,\nu'j'})]$$
$$\times[\sqrt{\sigma_j^c}\sqrt{\sigma_{j'}^c}+2i\bar{n}\sqrt{\sigma_j^c}\sqrt{\tau_{j'}^c}+\sqrt{\tau_j^c}\sqrt{\tau_{j'}^c}L_{\nu\nu'}(t)+\delta_{\nu\nu'}\delta_{jj'}(\sigma_j^{inc}+\tau_j^{inc}L_{\nu\nu}(t))]$$
$$\times\langle\exp[-i\mathbf{q}\mathbf{u}_{\nu j}(0)]\exp[i\mathbf{q}\mathbf{u}_{\nu'j'}(t)]\rangle+4\pi\sum_\nu\sigma_\nu^M(\mathbf{q})L_{\nu\nu}(t)$$
$$\times\langle\exp[-i\mathbf{q}\mathbf{u}_\nu(0)]\exp[i\mathbf{q}\mathbf{u}_\nu(t)]\rangle\Big\}+\text{H.c.},\qquad(11)$$

where $E=E_{p_1}-E_{p_2}$ is the energy transferred by the neutron, $\bar{n}=\bar{N}/\mathfrak{N}$ is the average density of the excitons in the crystal. The angle brackets in (11) denote statistical averaging over the Gibbs canonical ensemble. The mean value $L_{\nu\nu'}(t)\equiv\langle N_\nu(0)N_{\nu'}(t)\rangle$ is defined in terms of the exciton density-density correlation function:

$$L_{\nu\nu'}(t)\approx\bar{n}^2+\mathfrak{N}^{-1}\sum_{\mathbf{k}\neq0}\langle\rho_\mathbf{k}(0)\rho_{-\mathbf{k}}(t)\rangle\exp[i\mathbf{k}(\mathbf{R}_{0,\nu}-\mathbf{R}_{0,\nu'})].\qquad(12)$$

The first term in (12) yields the contribution to the elastic-scattering cross section; the second term, which depends on the time and describes the correlations of the electronic excitations, enter in the inelastic-scattering cross section. In the limit of zero electron-excitation density, Eq. (11) goes over into the formula for the cross section for nuclear scattering in the absence of excitons.[20]

Inelastic neutron scattering at transfer energies lower than the width of the exciton band is connected with the intraband exciton transitions. The expression for the inelastic-scattering cross section will be obtained on the basis of Eqs. (11) and (12):

$$\frac{\delta^2\Sigma^{ex}}{\delta E\delta\Omega}=\frac{1}{8\pi^2}\frac{p_2}{p_1}\sum_{\mathbf{k}\neq0}\int_{-\infty}^{\infty}dt\,e^{-iEt}\langle\rho_\mathbf{k}(0)\rho_{-\mathbf{k}}(t)\rangle$$
$$\times\Big\{\Lambda^c(\mathbf{q})\frac{(2\pi)^3}{v_0}\sum_\mathbf{g}\delta(\mathbf{q}-\mathbf{k}-2\pi\mathbf{g})+\Lambda^{inc}(\mathbf{q})\Big\}.\qquad(13)$$

Here $\mathbf{g}$ is the reciprocal-lattice vector and v_0 is the unit-cell volume. In (13) we have also introduce the notation

$$\Lambda^c(\mathbf{q})=\Big|\sum_j\sqrt{\tau_j^c}\exp(-i\mathbf{q}\mathbf{R}_{0,j})\exp(-W_j(\mathbf{q}))\Big|^2,\qquad(14)$$

$$\Lambda^{inc}(\mathbf{q})=\sum_j\tau_j^{inc}\exp(-2W_j(\mathbf{q}))+4\pi\sigma^M(\mathbf{q})\exp(-2W(\mathbf{q})),\qquad(15)$$

where $W_j(\mathbf{q})$ and $W(\mathbf{q})$ are the Debye-Waller factors[19,20]; $\Lambda^c(\mathbf{q})$ and $\Lambda^{inc}(\mathbf{q})$ pertain to coherent and incoherent scattering, respectively.

The cross section (13) is proportional to the dynamic structure factor of the exciton system, a factor defined as the Fourier transform of the correlation function $\langle\rho_k(0)\rho_{-k}(0)\rangle$. The latter, naturally, appears within the framework of the operator approach used to write down the matrix elements (9) and (10). The terms containing τ_j in Eqs. (13)–(15) correspond to neutron-exciton interaction connected with the change of the nuclear configuration, while the term with $\sigma^M(\mathbf{q})$, which appears in the case of triplet excitons, corresponds to the magnetic interaction. Both types of interaction lead to excitonic transitions because of the spatial correlations in the system of molecular excitons.

The correlation function that enters in (14) was obtained by us[21] by the method of equal-time Green's functions[19,22] with allowance for the exact commutation relations (3). We present for it an expression at $k\neq0$ in the Tyablikov approximation:

$$\langle\rho_\mathbf{k}(0)\rho_{-\mathbf{k}}(t)\rangle=\mathfrak{N}^{-1}\sum_\mathbf{p}\{\bar{n}_\mathbf{p}+(2\sigma)^{-1}\bar{n}_\mathbf{p}\bar{n}_{\mathbf{p}+\mathbf{k}}-2\bar{n}M_{\mathbf{p},\mathbf{p}+\mathbf{k}}\}\exp[i(\varepsilon_{\mathbf{p}+\mathbf{k}}^T-\varepsilon_\mathbf{p}^T)t],\qquad(16)$$

where $2\sigma=1-2\bar{n}$ is a parameter analogous to the order parameter of magnetism theory,[19]

$$\varepsilon_\mathbf{p}^T=\Delta+2\bar{n}F_0+2\sigma Q_\mathbf{p}\qquad(17)$$

is the exciton spectrum in the considered approximation,[15]

$$\bar{n}_\mathbf{p}=2\sigma\{\exp((\varepsilon_\mathbf{p}^T-\mu)/k_BT)-1\}^{-1}\qquad(18)$$

is the distribution function in the quasimomenta. The quantity $M_{\mathbf{p},\mathbf{p}+k}$ is given by

$$M_{\mathbf{p},\mathbf{p}+\mathbf{k}}=\frac{1-\alpha}{k_BT}\frac{Q_{\mathbf{p}+\mathbf{k}}-Q_\mathbf{p}}{\bar{n}_\mathbf{p}-\bar{n}_{\mathbf{p}+\mathbf{k}}}\overline{(\Delta n_\mathbf{p})^2}\,\overline{(\Delta n_{\mathbf{p}+\mathbf{k}})^2},\qquad(19)$$

where $\overline{(\Delta n_\mathbf{p})^2}=\bar{n}_\mathbf{p}[1+(\frac{1}{2}\sigma)\bar{n}_\mathbf{p}]$ is the mean squared fluctuation of the number of particles in state $\mathbf{p}$, and α is a splitting parameter similar to that introduced in Ref. 23. The value of α is chosen to satisfy the sum rule

$$\sum_\mathbf{k}\langle\rho_\mathbf{k}(0)\rho_{-\mathbf{k}}(0)\rangle=\bar{N}.\qquad(20)$$

As $\bar{n}\to0$ formula (16) goes over into the formula for the correlation function of an ideal Bose gas.[24]

The cross section for the magnetic scattering by excitons with $S=1$, obtained with allowance for the explicit form of the correlator (16), is

$$\frac{\delta^2\Sigma^M}{\delta E\delta\Omega}=\frac{p_2}{p}\sigma^M(\mathbf{q})e^{-2W(\mathbf{q})}\mathfrak{N}^{-1}\sum_{\mathbf{k}\neq0}\sum_\mathbf{p}\{\bar{n}_\mathbf{p}+(2\sigma)^{-1}\bar{n}_\mathbf{p}\bar{n}_{\mathbf{p}+\mathbf{k}}-2\bar{n}M_{\mathbf{p},\mathbf{p}+\mathbf{k}}\}$$
$$\times\delta(E-\varepsilon_{\mathbf{p}+\mathbf{k}}^T+\varepsilon_\mathbf{p}^T).\qquad(21)$$

Let us estimate the cross sections of the inelastic nuclear and magnetic scattering by excitons, using Eqs.

(13) and (21). Since we are interested in intraband transitions, we consider a neutron energy transfer $E \approx 10^{-4}-10^{-1}$ eV. The shift of the equilibrium positions of the nuclei in electronic excitations of aromatic molecules in a crystal is of the order of $\Delta R_{0,j} \sim 0.1$ Å.[25] For $E \sim 0.1$ eV this leads to $\tau_j \sim 0.1\ \sigma_j$, and for the limiting concentration $\bar{n} \sim 5\times10^{-3}$ the nuclear channel of elastic scattering by excitons in a wide-band crystal amounts to $10^{-4}-10^{-3}$ of the total nuclear elastic-scattering cross section. Since τ_j is proportional to E, nuclear scattering by excitons can be neglected in the case of very slow neutrons. The magnetic scattering is determined by the value of the form factor $\chi(\mathbf{q})$, which at $q > 10^8$ cm^{-1} decreases rapidly in accord with a power law. In the case of very small energy transfers, $E < 10^{-4}$ eV we have $\chi(q) \approx \chi(0) = 1$ and the cross section for magnetic scattering by excitons is $\sim 0.4 n$ [b] per crystal molecule, i.e., at an exciton density $\bar{n} \sim 5\times10^{-3}$ it equals several millibarns. At $E > 10^{-3}$ eV the magnetic scattering is practically nonexistent because of the smallness of the form factor; at these energies only nuclear scattering by the excitons is significant.

3. INELASTIC SCATTERING OF NEUTRONS IN THE PRESENCE OF AN EXCITON CONDENSATE

The possibility of Bose condensation of molecular excitons was demonstrated in Ref. 15. In this section we consider the effect of formation of a Bose-condensed state of excitons on the process of inelastic scattering of neutrons in a crystal.

Besides the inelastic scattering channels considered above, we shall take into account magnetic scattering accompanied by creation and annihilation of excitons. The form factor of the inelastic scattering decreases at large q more rapidly than $(\rho q)^{-8}$, where ρ is the average radius of the electronic state.[7] Therefore magnetic scattering with excitation of an exciton is significant at high neutron energies ~10^3 meV and at low values $q \lesssim 10^8$ cm^{-1}, whereas at low energies they can be neglected. The additional channel of scattering with change of the number of excitons will be taken into account with the aid of the approach proposed by Fedyanin and Yakushevich.[8] To this end we represent the matrix element $V(\mathbf{q})$ in the form of an expansion in the operators of creation and annihilation of the electronic excitations, b_ν^+ and b_ν:

$$V(\mathbf{q}) = \sum_\nu (a_\nu(\mathbf{q}) + a_{\downarrow\nu}(\mathbf{q}) b_\nu + a_{\uparrow\nu}(\mathbf{q}) b_\nu^+ + \Delta a_\nu(\mathbf{q}) N_\nu) \exp(i\mathbf{q}\mathbf{R}_\nu), \tag{22}$$

where a_ν and Δa_ν are defined in Eqs. (7) and (8), while $a_{\downarrow\nu}$ and $a_{\uparrow\nu}$ are equal in the case of magnetic interactions to

$$a_{\uparrow\nu}(\mathbf{q}) = a_{\downarrow\nu}^*(-\mathbf{q}) = -\frac{4\pi r_e \gamma_n}{m_n} \langle f| \sum_i \mathbf{s}_n [\mathbf{e}\times[\boldsymbol{\mu}_i\times\mathbf{e}]] e^{i\mathbf{q}\mathbf{r}_i} |0\rangle, \tag{23}$$

$\mathbf{r}_i$ is the coordinate of the i-th electron, μ_i is expressed in terms of the spin operator $\mathbf{s}_i$ and the momentum operator $\mathbf{p}_i$ of the electron:

$$\boldsymbol{\mu}_i = \mathbf{s}_i - [\mathbf{e}\times\mathbf{p}_i] i/q.$$

The summation in (23) is over all the electrons of the molecule of cell ν.

The calculation of the cross section for the inelastic scattering for the potential (22) is similar to the calculation of the cross section (13) with potential (9). For the coherent part of the scattering we have

$$\frac{\delta^2\Sigma^c}{\delta E\delta\Omega} = \mathfrak{R}\frac{1}{8\pi^2}\frac{p_2}{p_1}\int_{-\infty}^{\infty} dt\, e^{-iEt}\{\Lambda_\uparrow(\mathbf{q})\langle a_\mathbf{k}(0)a_\mathbf{k}^+(t) + a_{-\mathbf{k}}^+(0)a_{-\mathbf{k}}(t)\rangle$$

$$+\Lambda^c(\mathbf{q})(1-\delta_{\mathbf{k}0})\langle\rho_\mathbf{k}(0)\rho_{-\mathbf{k}}(t)\rangle\}, \quad \mathbf{k}=\mathbf{q}-2\pi\mathbf{g}, \tag{24}$$

where $\Lambda^c(\mathbf{q})$ is defined by (14), and $\Lambda_\uparrow(\mathbf{q})$ is given by

$$\Lambda_\uparrow(\mathbf{q}) = \Lambda_\downarrow(\mathbf{q}) = 4\pi r_e^2\gamma_n^2 \exp(-2W(\mathbf{q}))$$
$$\times|\langle f|\sum_i [\mathbf{e}[\boldsymbol{\mu}_i, \mathbf{e}]]\exp(i\mathbf{q}\mathbf{r}_i)|0\rangle|^2. \tag{25}$$

The term with $\Lambda^c(\mathbf{q})$ in (24) corresponds to nuclear scattering by excitons, while the term with $\Lambda_\uparrow(\mathbf{q})$ corresponds to magnetic scattering with change of the number of excitons. Equation (24) was obtained in the approximation $\langle a_k a_{-k}\rangle = \langle a_{-k}^+ a_k^+\rangle = 0$.

We now take into account the presence in the condensate of a macroscopic number N_0 of quasiparticles in a state with $\mathbf{k}=0$. We shall assume that N_0 satisfies the condition $N_0 \gg N - N_0$ (N is the total number of excitons). We perform in (24) the shift operation on the excitonic operators[26]

$$a_\mathbf{k}(t) = \sqrt{N_0^*}e^{-i\mu t}\delta_{\mathbf{k}0} + c_\mathbf{k}(t). \tag{26}$$

Since the excitons do not have Bose statistics, Eq. (26) contains in place of the number N_0 of excitons in the condensate the quantity N_0^*[15]:

$$N_0^* \approx N_0(1 - N_0/\mathfrak{R}). \tag{27}$$

We introduce the operators of the above-condensate elementary excitations $\tilde{c}_k^+$ and $\tilde{c}_k$,[15,16] which are connected with the operators c_k^+ and c_k by the transformations

$$c_\mathbf{k} = u_\mathbf{k}\tilde{c}_\mathbf{k} + v_\mathbf{k}\tilde{c}^+_{-\mathbf{k}}, \quad c_\mathbf{k}^+ = u_\mathbf{k}\tilde{c}_\mathbf{k}^+ + v_\mathbf{k}\tilde{c}_{-\mathbf{k}}. \tag{28}$$

The transformation coefficients in (28) are

$$u_\mathbf{k} = \left[\frac{1}{2}\left(1+\frac{\xi_\mathbf{k}}{\bar{\varepsilon}_\mathbf{k}}\right)\right]^{1/2}, \quad v_\mathbf{k} = \left[\frac{1}{2}\left(-1+\frac{\xi_\mathbf{k}}{\bar{\varepsilon}_\mathbf{k}}\right)\right]^{1/2}; \tag{29}$$

$$\xi_\mathbf{k} = Q_\mathbf{k} - Q_0 + 2n_0^*(F_\mathbf{k} - Q_\mathbf{k}), \quad n_0^* = N_0^*/\mathfrak{R} = {}^1/_2 - \sigma_0^*;$$
$$\eta_\mathbf{k} = 2n_0^*(F_\mathbf{k} - Q_0); \tag{30}$$

$$\bar{\varepsilon}_\mathbf{k} = (\xi_\mathbf{k}^2 - \eta_\mathbf{k}^2)^{1/2} = 2[(Q_\mathbf{k}-Q_0)^2\sigma_0^{*2} + 2n_0^*\sigma_0^*(Q_\mathbf{k}-Q_0)(F_\mathbf{k}-Q_0)]^{1/2}; \tag{31}$$

$\bar{\varepsilon}_k$ is the energy spectrum of the elementary excitations with a distribution function

$$\tilde{n}_\mathbf{k} = \langle\tilde{c}_\mathbf{k}^+\tilde{c}_\mathbf{k}\rangle = 2\sigma_0^*(\exp(\bar{\varepsilon}_\mathbf{k}/k_B T) - 1)^{-1}. \tag{32}$$

The final form for the coherent-scattering cross section, obtained with (28)–(31) taken into account, is

$$\frac{\delta^2\Sigma^c}{\delta E\delta\Omega}(\mathrm{cond}) = \mathfrak{R}\frac{1}{4\pi}\frac{p_2}{p_1}\Big\{N_0^*\delta_{\mathbf{k}0}\Lambda_\uparrow(\mathbf{q})(\delta(E-\mu)+\delta(E+\mu))$$
$$+\frac{\xi_\mathbf{k}}{\bar{\varepsilon}_\mathbf{k}}\Lambda_\uparrow(\mathbf{q})[(2\sigma_0^*+\tilde{n}_\mathbf{k})\delta(E-\mu-\bar{\varepsilon}_\mathbf{k}) + \tilde{n}_\mathbf{k}\delta(E+\mu+\bar{\varepsilon}_\mathbf{k})]$$
$$+n_0^*(1-\delta_{\mathbf{k}0})\Lambda^c(\mathbf{q})\Big[\frac{1}{2}\Big(1+\frac{\xi_\mathbf{k}-\eta_\mathbf{k}}{\bar{\varepsilon}_\mathbf{k}}\Big)((2\sigma_0^*+\tilde{n}_\mathbf{k})\delta(E-\bar{\varepsilon}_\mathbf{k}) + \tilde{n}_\mathbf{k}\delta(E+\bar{\varepsilon}_\mathbf{k}))$$
$$+\frac{1}{2}-1+\frac{\xi_\mathbf{k}-\eta_\mathbf{k}}{\bar{\varepsilon}_\mathbf{k}}((2\sigma_0^*+\tilde{n}_\mathbf{k})\delta(E-2\mu-\bar{\varepsilon}_\mathbf{k}) + \tilde{n}_\mathbf{k}\delta(E+2\mu+\bar{\varepsilon}_\mathbf{k}))\Big]\Big\}. \tag{33}$$

The quasimomentum $\mathbf{k}$ satisfies the condition $\mathbf{k} = \mathbf{q} - 2\pi\mathbf{g}$. The first term in the curly brackets of (33) corresponds to creation or annihilation of an exciton in the condensate with energy μ. The second term corresponds to creation (annihilation) of a condensate quasiparticle μ and an above-condensate gapless elementary excitation $\tilde{\varepsilon}_k$. In the absence of a Bose condensate this term corresponds to production of an exciton with energy ε_k, and in this case $\mu + \tilde{\varepsilon}_k$ in the argument of the δ-function in (33) is replaced by ε_k. The third term in (33) corresponds to creation (annihilation) of only above-condensate excitation, with the macroscopic number of excitons in the condensate remaining constant in the scattering process (the analog of the creation of elementary excitations in the scattering of neutrons in liquid He). This term is connected with the collective properties of the interacting excitons produced by intense laser pumping. The last term in the curly brackets describes the simultaneous creation (annihilation) of a pair of excitons in the condensate and of one elementary excitation. Since the number of the above-condensate elementary excitations is small, the transitions between them are not taken into account here.

As follows from (33), the part of the inelastic scattering connected with the absorption and creation of an exciton in a Bose condensate has two δ-like peaks. Observation of sharp maxima of equal intensity in inelastic scattering, corresponding to

$$E=\pm\mu, \quad \mathbf{q}=2\pi\mathbf{g}, \tag{34}$$

would provide experimental evidence of the presence of the Bose condensate and permit the determination of the number of excitons in it. Inasmuch as in magnetic excitation of a condensate quasiparticle the value of q must be small, the condition (34) is satisfied by vectors $2\pi\mathbf{g}$ lying in the first Brillouin zone.

To estimate the terms connected with the production of elementary excitations, we put $q \sim 10^8$ cm^{-1}. The form factor in terms of which $\Lambda_+(q)$ (25) is expressed has in this case a value not lower than $10^{-1}-10^{-2}$. Putting $p_2 \sim p_1$ and $\xi_k \sim \tilde{\varepsilon}_k$, we obtain for the cross section with production of an above-condensate excitation $(10^{-2}-10^{-3})$ $\mathfrak{N}$ [b].

4. ROLE OF EXCITON-PHONON INTERACTION IN THE NEUTRON SCATTERING PROCESS

Allowance for the electron-vibrational interaction leads to a mixing of the electron and nuclear motions. In quasiparticle language this means a renormalization of the exciton and phonon states upon diagonalization of the initial Hamiltonian. The result is a nonzero probability of exciting excitons in the course of nuclear inelastic neutron scattering.[5] It was noted in the Introduction that calculation of the cross section of similar transitions in Refs. 5–7 led to discrepancies in the estimates of the cross sections and of their dependences on the exciton energy. In this section we use expressions for the cross sections of excitonic transitions in neutron scattering, using a Hamiltonian with an exciton-phonon interaction and containing terms that are linear and quadratic in the exciton operators. We assume for simplicity that the excitons interact with one optical branch of the phonons

$$H=H_{ex}+H_{ph}+H_{ex-ph}, \tag{35}$$

where H_{ex} is given by expression (1) with $\mu=0$,

$$H_{ph}=\sum_{\mathbf{k}}\omega_{\mathbf{k}}b_{\mathbf{k}}^{+}b_{\mathbf{k}}, \tag{36}$$

$$H_{ex-ph}=\sum_{\mathbf{k}}\gamma_{\mathbf{k}}(a_{\mathbf{k}}^{+}+a_{-\mathbf{k}})\varphi_{\mathbf{k}}+\mathfrak{N}^{-1/2}\sum_{\mathbf{pk}}G_{\mathbf{pk}}a_{\mathbf{p+k}}^{+}a_{\mathbf{p}}\varphi_{\mathbf{k}}. \tag{37}$$

We have introduced in (37) the symbols $\varphi_k = b_k + b_{-k}^{+}$; G_{pk} and γ_k are the constants of the exciton-phonon interactions with and without conservation of the number of excitons in the scattering. The constant γ_k for intramolecular vibrations is of the order of[5,6] $\gamma \sim 0.1$ eV. The second term in the interaction operator (37) corresponds to the approximation of a weak exciton-phonon coupling.[17] The constant G_{pk} for the internal vibrations is easiest to estimate at small values of p and k at[27] $G \sim V_0 a^{-1}(2m\omega_0)^{-1/2}$, where a is the lattice period and V_0 is the width of the exciton band. For crystal with a bandwidth ~0.1 eV we obtain $G \sim 10^{-2}$ eV. Since the interaction γ, which is not diagonal in the number of excitons, does not lead to damping of the exciton and phonon states, it can be treated as a renormalization of the excitons and the phonons. The diagonal interaction G will describe the scattering processes. The quasiparticle damping associated with it should lead to a smearing of the energy distribution of the inelastically scattered neutrons.[28] This manifests itself in the replacement of the δ-functions in the formulas for the cross sections by levels of finite width.

To find the inelastic-scattering cross section we expand the displacement $\mathbf{u}_{\nu j}$ of the atoms in the phonon operators:

$$\mathbf{u}_{\nu j}=\mathfrak{N}^{-1/2}\sum_{\mathbf{k}}\mathbf{u}_j(\mathbf{k})\,(b_{\mathbf{k}}+b_{-\mathbf{k}}^{+})\exp(i\mathbf{k}\mathbf{R}_{0,\nu}) \tag{38}$$

and use the procedure described in Ref. 5, representing the sum $b_k^{+} b_{-k}^{+}$ in (38) in terms of the operators of the renormalized quasiparticles. The procedure for the renormalization of the phonon and exciton operators, necessitated by the off-diagonal exciton-phonon interaction, should be carried out in our approximation with allowance for the fact that the excitons do not have Bose statistics. By solving the equations of motion for the operators a_k and b_k with the commutation relations (3) under the condition γ_k, $\omega_k \ll \varepsilon_k$, we obtain

$$b_{\mathbf{k}}+b_{-\mathbf{k}}^{+}\approx\beta_{\mathbf{k}}+\beta_{-\mathbf{k}}^{+}+\frac{2\omega_{\mathbf{k}}\gamma_{\mathbf{k}}}{E_{\mathbf{k}}^{2}-\omega_{\mathbf{k}}^{2}}(\alpha_{\mathbf{k}}+\alpha_{-\mathbf{k}}^{+}), \tag{39}$$

where the energy of the "dressed" excitons E_k is determined by the equality

$$E_{\mathbf{k}}\approx\varepsilon_{\mathbf{k}}^{T}\left(1+\frac{4\sigma\omega_{\mathbf{k}}|\gamma_{\mathbf{k}}|^{2}}{(\varepsilon_{\mathbf{k}}^{T})^{3}}\right),$$

ε_k^T is the energy of the excitons without allowance for the exciton-phonon interaction in the Tyablikov approximation (17).

We introduce for future use the quantities

$$\Pi^{c}(\mathbf{k},\mathbf{q})=\Big|\sum_{j}\sqrt{\sigma_j^{c}}\,\mathbf{q}\mathbf{u}_j(\mathbf{k})\,e^{-i\mathbf{q}\mathbf{R}_{0,j}}e^{-\bar{W}_j(\mathbf{q})}\Big|^{2}, \tag{40}$$

$$\Pi^{inc}(\mathbf{k},\mathbf{q})=\sum_{j}\sigma_j^{inc}\,|\mathbf{q}\mathbf{u}_j(\mathbf{k})|^{2}\,e^{-2\bar{W}_j(\mathbf{q})}, \tag{41}$$

$\tilde{W}_j(\mathbf{q})$ in (40) and (41) is the Debye-Waller factor for the renormalized phonons:

$$\tilde{W}_j(\mathbf{q})=\frac{1}{2\mathfrak{N}}\sum_{\mathbf{k}}|\mathbf{q}\mathbf{u}_j(\mathbf{k})|^2(1+2\langle\beta_{\mathbf{k}}^+\beta_{\mathbf{k}}\rangle). \quad (42)$$

Using formulas (12), (38), and (39), we express the scattering cross sections (11) in the presence of excitons in terms of the correlation functions of the exciton operators. The final expressions for the cross sections of the coherent and incoherent scattering of neutrons by a system of excitons are

$$\frac{\delta^2\Sigma^c}{\delta E\delta\Omega}=\mathfrak{N}\frac{1}{4\pi}\frac{p_2}{p_1}F^c(\mathbf{k},\mathbf{q},E),\quad \mathbf{k}=\mathbf{q}-2\pi\mathbf{g}. \quad (43)$$

$$\frac{\delta^2\Sigma^{inc}}{\delta E\delta\Omega}=\frac{1}{4\pi}\frac{p_2}{p_1}\sum_{\mathbf{k}}F^{inc}(\mathbf{k},\mathbf{q},E), \quad (44)$$

where the function $F^c(\mathbf{k},\mathbf{q},E)$ is expressed in terms of $\Lambda^c(\mathbf{q})$ (14) and $\Pi^c(\mathbf{k},\mathbf{q})$ (40):

$$F^c(\mathbf{k},\mathbf{q},E)=\frac{4\omega_{\mathbf{k}}^2|\gamma_{\mathbf{k}}|^2}{(E_{\mathbf{k}}^2-\omega_{\mathbf{k}}^2)^2}\Pi^c(\mathbf{k},\mathbf{q})\{(2\sigma+\bar{n}_{\mathbf{k}})\zeta(\Gamma_1;E-E_{\mathbf{k}}-\Delta_1)+\bar{n}_{\mathbf{k}}\zeta(\Gamma_1;E+E_{\mathbf{k}}+\Delta_1)\}+\Lambda^c(\mathbf{q})(1-\delta_{\mathbf{k}0})(2\sigma\mathfrak{N})^{-1}\times\sum_{\mathbf{p}}(2\sigma+\bar{n}_{\mathbf{p}+\mathbf{k}})\bar{n}_{\mathbf{p}}\zeta(\Gamma_2;E-E_{\mathbf{p}+\mathbf{k}}+E_{\mathbf{p}}-\Delta_2), \quad (45)$$

Δ_1, Δ_2, and Γ_1, Γ_2 denote the real and imaginary parts of the mass operators for the one-exciton and two-exciton Green's functions, respectively. The quantity $F^{inc}(\mathbf{k},\mathbf{q},E)$ in (44) is obtained from $F^c(\mathbf{k},\mathbf{q},E)$ (45) by making the substitutions $\Lambda^c\to\Lambda^{inc}$ and $\Pi^c\to\Pi^{inc}$. The functions ζ in (45) are equal to

$$\zeta(x;\,y)=\pi^{-1}x/(y^2+x^2) \quad (46)$$

and go over into $\delta(y)$ as $x\to 0$. The function $\zeta(\Gamma_1;E-E_k-\Delta_1)$, defined in terms of the exciton band shift Δ_1 and the exciton damping Γ_1, coincides at $k=0$ with the form function of the exciton-absorption band.[29] The first term with the coefficient $\Pi^c(\mathbf{k},\mathbf{q})$ in (45) describes zero-phonon creation and annihilation of excitons. The second term, which contains $\Lambda^c(\mathbf{q})$, corresponds to intraband transitions in the scattering of neutrons. At zero exciton density and when the functions of the widths of the exciton levels (46) are replaced by δ-functions, expressions (43)–(45) are equivalent to the corresponding formulas of the paper of Krivoglaz and Rashba.[6]

The cross section for scattering with production of an exciton of energy E_k is proportional to E_k^{-4} at $E_k \gg \omega_k$. A similar dependence on the energy of the electronic transition is obtained from calculations using the nonadiabaticity operator.[6,9,30] Agranovich and Lalov[5] obtained an exciton-production cross section proportional to $(mn/M)E_k^{-3}$. Such a dependence can be obtained from the formulas of the present article in the particular case $p_2 \ll p_1$ if we put $q^2\approx 2m_nE_k$. However, the estimates given in Ref. 5 for the ratio of the exciton production cross section to the phonon production cross sections are overvalued; the conclusion in Ref. 5 that the mechanism of excitation of excitons by neutrons differs from the mechanism of excitation of molecular states does not follow from our analysis.

Elliott and Shukla[7] obtained a cross section $\propto E_k^{-2}$. The reason for obtaining such a dependence is failure to take into account the exciton-phonon interaction for the ground state. When this interaction is consistently taken into account the terms proportional to γ_k/E_k in the matrix element of the transition cancel out in the principal order. As a result, the matrix element shows an E_k^{-2} dependence and the cross section an E_k^{-4} dependence.

For that part of the cross section which corresponds to the first term in (45), the estimates of Ref. 6 are applicable. The contribution from the second term at $E\sim 0.1$ eV amounts to $\sim 0.1\bar{n}_k$ of the differential cross section of the elastic nuclear scattering ($\bar{n}_k$ are the occupation numbers of the states in which the exciton is annihilated).

In conclusion, we consider inelastic neutron scattering in the presence of an exciton Bose condensate, due to exciton-phonon interaction. We shall take into account only the diagonal part of the exciton-phonon interaction, assuming that the nondiagonal interaction can be reduced if necessary to a renormalization of the initial exciton and phonon states,

$$H=\sum_{\mathbf{k}}(\varepsilon_{\mathbf{k}}-\mu)a_{\mathbf{k}}^+a_{\mathbf{k}}+\sum_{\mathbf{k}}F_{\mathbf{k}}\rho_{\mathbf{k}}\rho_{-\mathbf{k}}+\mathfrak{N}^{-1/2}\sum_{\mathbf{p}\mathbf{k}}G_{\mathbf{p}\mathbf{k}}a_{\mathbf{p}+\mathbf{k}}^+a_{\mathbf{p}}\varphi_{\mathbf{k}}. \quad (47)$$

The Bose condensation of the excitons, described by a Hamiltonian of the type (47) with Bose operators a_k, was considered by Moskalenko.[13]

The shift transformation (26) separates, in the exciton-phonon interaction operator in (47), the terms that are quadratic in the exciton and phonon operators and are proportional to $(n_0^*)^{1/2}$. We take them into account by renormalizing the spectrum of the above-condensate excitations, assuming that the dispersion of the initial above-condensate excitations is small, i.e., for k lying in the first Brillouin zone, the following inequality holds:

$$\max\tilde{\varepsilon}_{\mathbf{k}}<\omega_{\mathbf{k}}, \quad (48)$$

where ω_k is the frequency of the optical phonon and $\tilde{\varepsilon}_k$ is given by formula (31). The energy of the renormalized above-condensate excitations $\tilde{E}_k$ is obtained from the formula

$$\tilde{E}_{\mathbf{k}}^2={}^1/_2(\tilde{\varepsilon}_{\mathbf{k}}^2+\omega_{\mathbf{k}}^2)+{}^1/_2[(\tilde{\varepsilon}_{\mathbf{k}}^2-\omega_{\mathbf{k}}^2)^2+32n_0^*\sigma_0^*\omega_{\mathbf{k}}\tilde{\varepsilon}_{\mathbf{k}}|\tilde{G}_{0\mathbf{k}}|^2]^{1/2}. \quad (49)$$

Inelastic neutron scattering should be connected with creation and annihilation of above-condensate elementary excitations. Owing to the low density of the above-condensate excitations, the transitions between them can be neglected. The coherent-scattering cross section is given by

$$\frac{\delta^2\Sigma^c}{\delta E\delta\Omega}(\text{cond})\approx N_0^*\frac{1}{\pi}\frac{p_2}{p_1}\frac{\omega_{\mathbf{k}}^2|\tilde{G}_{0\mathbf{k}}|^2}{(\tilde{E}_{\mathbf{k}}^2-\omega_{\mathbf{k}}^2)^2}\Pi^c(\mathbf{k},\mathbf{q})\times\{(2\sigma_0^*+\bar{n}_{\mathbf{k}})\delta(E-\tilde{E}_{\mathbf{k}})+\bar{n}_{\mathbf{k}}\delta(E+\tilde{E}_{\mathbf{k}})\}, \quad (50)$$

$$\tilde{G}_{0\mathbf{k}}=G_{0\mathbf{k}}\left(\frac{\xi_{\mathbf{k}}-\eta_{\mathbf{k}}}{\xi_{\mathbf{k}}+\eta_{\mathbf{k}}}\right)^{1/2}. \quad (51)$$

The distribution function $\bar{n}_k$ in (50) is expressed in terms of the energy $\tilde{E}_k$ (49) in analogy with $\bar{n}_k$ in (32).

The cross section (50) per lattice site is determined by the characteristic factor

$$n_0^*\omega_{\mathbf{k}}^2|\tilde{G}_{0\mathbf{k}}|^2/(\tilde{E}_{\mathbf{k}}^2-\omega_{\mathbf{k}}^2)^2,$$

which has, accurate to a coefficient n_0^* the same structure as in the considered case of scattering with production of an exciton in the absence of a condensate. Since $\bar{E}_k < \omega_k$, this factor can become $\sim n_0^*$ for low-frequency optical phonons (orientational vibrations of the molecules). The cross section for the transition of the particle into an above-condensate state can accordingly take on values ($10^{-1}-10^{-2}$) of the nuclear scattering cross section. Inasmuch as the quasimomentum of the elementary excitation is equal, accurate to a vector $2\pi \mathbf{g}$, to the transferred momentum $\mathbf{q}$, in the considered processes in which a condensate quasiparticle takes part, the study of the spectrum of thermal neutrons in inelastic scattering may serve as a method of determining the dispersion law of elementary above-condensate excitations E_k.

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