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MANY-PHONON PROCESSES IN CRYSTALS

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We have calculated the differential cross section for the inelastic scattering of a neutron by a crystal with either emission or absorption of an arbitrary number of phonons. The first case is of interest when the temperature of the crystal is low, and the second when the neutron energy is very small and the crystal temperature sufficiently high. Formulae are obtained for crystals with an arbitrary spectrum of the normal vibrations. If we choose a specific spectrum, the calculations can be pursued to the end, leading to a simple final formula. We have also given the formula for the limiting case of the scattering of high energy neutrons by a free nucleus.

1. INTRODUCTION

To investigate the interaction of slow neutrons with crystalline substances, one uses the Debye model of a crystal. In that case the transfer of energy from the neutron to the crystal is treated as the excitation of one or several "phonons," that is, quanta of the thermal motion of the crystal. The transfer of energy from the crystal to the neutron corresponds to the absorption of phonons by the neutron. These processes have often been considered in the literature. Weinstock¹ derived formulae for the effective cross section for elastic and inelastic neutron scattering with the emission or absorption of one phonon (one-phonon process). The evaluation of processes involving simultaneously the emission and absorption of several phonons is in principle not difficult, but in practice very cumbersome.

Squires² in calculating the cross section for scattering of slow neutrons by Mg and Ni considered terms d_{mn} corresponding to processes where m phonons are emitted and n phonons absorbed. For $m + n \geq 2$ he did not take into account the interference between the waves scattered coherently from different atoms. Squires' calculations agree well with his own experiments. The neutron energy in those experiments was very small (≈ 0.003 ev), and it was therefore sufficient for the author to calculate several terms with the smallest values of m and n . In those cases where the number of phonons involved in the scattering can be large, the number of terms d_{mn} contributing to the cross section also becomes large; it becomes therefore impossible to evaluate the cross section by evaluating every term separately, as was done by Squires, and it is necessary to develop a method for summing the terms d_{mn} .

In the present paper we calculate the cross section for inelastic neutron scattering by evaluating only processes of identical character: either only emission, or only absorption of an arbitrary number of phonons. In Squires' notation this corresponds to $\sum_{m=1}^{\infty} d_{m0}$ and $\sum_{n=1}^{\infty} d_{0n}$. Interference is not considered.

The formulae obtained have practical value in two cases.

1. The case of low crystal temperatures and sufficiently large neutron energies. In this case absorp-

tion of phonons is unlikely, so that Σd_{m0} will give the total scattering cross section. (If the temperature of the crystal is not very low, one can evaluate from Weinstock's formula the cross section d_{01} for scattering involving the absorption of one phonon.) Furthermore, if the neutron energy is sufficiently large so that several Miller planes are involved in the inelastic scattering, interference can be neglected (we are dealing with polycrystalline matter). However, the formulae have an exact meaning, independent of the number of Miller planes involved; namely, if we take instead of $\overline{a^2}$ the amplitude of the incoherent scattering, $\overline{a^2} - (\overline{a})^2$, we obtain the incoherent part of the scattering of the neutron by the crystal. As the ratio E/Θ (Θ is the Debye temperature in energy units) tends to infinity, we find, as was to be expected, the formulae for the scattering of a neutron by a free nucleus.

2. The case of low neutron energies and sufficiently high crystal temperatures. In this case it is very unlikely that the neutron will lose energy; if the neutron wave length exceeds some critical value, the coherent part of elastic and inelastic scattering, involving loss of energy, will in general tend to zero, so that the sum Σd_{0n} will give the total inelastic scattering cross section. As regards interference, several Miller planes will be involved in the majority of substances even for zero neutron energy and the absorption of one phonon by the neutron.³ The number of planes involved increases rapidly with increasing number of absorbed phonons and increasing neutron energy.* Interference is thus of no consequence for practically important cases.

2. DERIVATION OF THE GENERAL FORMULAE

Since the theory of the scattering of slow neutrons by a crystal is well known (see, for instance, Refs. 1, 4, 5) we shall not discuss in detail the basic principles, but only remind our readers of them, and also explain the notation.

The scattering is considered in Born approximation; for the interaction between the neutron and the crystal we take the potential

$$V = \sum_{\mathbf{R}} \alpha_{\mathbf{R}} \delta(\mathbf{r} - \mathbf{R}). \quad (1)$$

where \mathbf{r} is the position of the neutron and \mathbf{R} that of the nucleus,

$$\alpha_{\mathbf{R}} = (2\pi\hbar^2/m)(1 + m/M_{\mathbf{R}})a_{\mathbf{R}}, \quad (2)$$

where m is the neutron mass, $M_{\mathbf{R}}$ the mass of the nucleus at the position \mathbf{R} , and $a_{\mathbf{R}}$ the scattering amplitude (depending in general on the nuclear spin). The sum is taken over all nuclei in the crystal.

As far as we are interested in the average cross section of scattering by one nucleus, assuming no interference to be present, we must leave in the sum (1) only one term corresponding to the interaction with the given nucleus $\mathbf{R} = \mathbf{R}_0$, and replace $\alpha_{\mathbf{R}}$ by α , defined by the equation $|\alpha|^2 = \overline{|\alpha_{\mathbf{R}}|^2}$, where the average is taken over the nuclei of the lattice and their spins,

$$V = \alpha \delta(\mathbf{r} - \mathbf{R}). \quad (1a)$$

Considering a crystal consisting of identical atoms of mass M and taking for M the average mass of the isotopes, one can easily obtain in Born approximation a formula for the differential cross section (with respect to angle and energy) for the scattering by one nucleus,

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k}{k_0} \overline{a^2} \left(1 + \frac{m}{M}\right)^2 \sum_{\tau} |F_{\tau\tau_0}|^2 \delta(E_{\tau} + E - E_{\tau_0} - E_0), \quad (3)$$

where \mathbf{k}_0 , E_0 , \mathbf{k} , and E are the wave vectors and energies of the neutron before and after scattering, τ_0 and τ the initial and final state of the crystal, and E_{τ_0} and E_{τ} the corresponding energies; $F_{\tau\tau_0}$ is the matrix element of the operator

$$F \equiv \exp \{i(\mathbf{k}_0 - \mathbf{k}) \cdot \mathbf{R}_0\}, \quad (4)$$

and $\overline{a^2}$ the square of the scattering amplitude averaged over the different isotopes and over the spins.

* For instance, for neutrons of wavelength 10.5 Å and scattering, involving one phonon by magnesium this number is 106 (Ref. 2).

We shall consider a crystal with a simple lattice and neglect the difference in mass of the various isotopes. We shall go over to the normal coordinates of the crystal using the equations

$$\mathbf{R} - \rho \equiv \mathbf{u}_\rho = \left(\frac{2}{N}\right)^{1/2} \sum_s \left\{ \sum_{j=1}^3 \mathbf{e}_{sj} \xi_{sj} B_{s\rho} \right\}, \quad (5)$$

$$B_{s\rho} = \begin{cases} \sin \mathbf{q}_s \rho, & \text{if } \mathbf{q}_s \in P_+ \\ \cos \mathbf{q}_s \rho, & \text{if } \mathbf{q}_s \in P_- \end{cases} \quad (6)$$

where ρ is the equilibrium position of the nucleus and \mathbf{u}_ρ the deviation from it. The vectors ρ form a spatial crystal lattice of N lattice points. Equation (5) is the expansion of \mathbf{u}_ρ in a Fourier series in the wave vectors \mathbf{q}_s which form the so-called reciprocal lattice which has also N lattice points, in wave vector space. This space we divide into two half-spaces by an arbitrary plane through the origin. The notation $\mathbf{q}_s \in P_+$ indicates that the end of the vector \mathbf{q}_s lies in a point of that half-space which we arbitrarily call the upper one, while $\mathbf{q}_s \in P_-$ indicates that \mathbf{q}_s belongs to the other (lower) half-space. The vectors \mathbf{e}_{sj} are three unit vectors corresponding to the polarization of the vibration; one of them is parallel and the other two are perpendicular to \mathbf{q}_s . N is the total number of nuclei in the crystal which tends to infinity. Finally, ξ_{sj} are the amplitudes of the expansion which also are the new (normal) coordinates of the crystal. The wave function of the crystal expressed in the new coordinates has the form

$$\Psi = \prod_{sj} \psi_{n_{sj}}(\xi_{sj}); \quad (7)$$

where ψ_n is the function of the linear oscillator in the n -th excited state. The operator F expressed in the new variables has the form

$$F = \exp \{i(\mathbf{k}_0 - \mathbf{k})(\rho_0 + \mathbf{u})\} = \exp \{i(\mathbf{k}_0 - \mathbf{k})\rho_0\} \prod_{sj} \exp \{i(\mathbf{k}_0 - \mathbf{k})\mathbf{e}_{sj}(2/N)^{1/2} B_{s\rho_0} \xi_{sj}\}. \quad (8)$$

If we put the origin in the point ρ_0 where the nucleus under consideration is situated and if we take into account that

$$B_{s0} = \begin{cases} 0, & \text{if } \mathbf{q}_s \in P_+ \\ 1, & \text{if } \mathbf{q}_s \in P_- \end{cases}$$

we get

$$F = \prod_{sj}^{P_-} \exp \{i(\mathbf{k}_0 - \mathbf{k})\mathbf{e}_{sj}(2/N)^{1/2} \xi_{sj}\}, \quad (9)$$

where the symbol P_- above the \prod indicates that only variables referring to the lower half-space enter into the product.

Thanks to the separation of variables in both the wave function and the operator the matrix element $F_{\tau\tau_0}$ can also be written as a product

$$F_{\tau\tau_0} = \prod_{sj}^{P_-} F_{n'_{sj}, n_{sj}}^{(sj)}, \quad (10)$$

where n_{sj} characterizes the initial state of the crystal and n'_{sj} its final state. It is easy to show¹ that as $N \rightarrow \infty$ the only final states that give a contribution different from zero are those for which n'_{sj} differs at most by unity from n_{sj} for each pair of indices (sj) , and that in the expansion of the corresponding matrix elements it is sufficient to take along only the dominant term for $N \rightarrow \infty$. These matrix elements have the form

$$F_{n'_{sj}, n_{sj}}^{(sj)} = 1 - [(\mathbf{k}_0 - \mathbf{k})\mathbf{e}_{sj}]^2 h \left(n + \frac{1}{2} \right) / NM\omega_{sj} + \dots, \quad (11)$$

$$F_{n_{sj}+1, n_{sj}}^{(sj)} = \frac{i}{\sqrt{2}} (\mathbf{k}_0 - \mathbf{k})\mathbf{e}_{sj} \left(\frac{2h}{NM\omega_{sj}} \right)^{1/2} (n_{sj} + 1)^{1/2} + \dots, \quad (12)$$

$$F_{n_{sj}-1, n_{sj}}^{(sj)} = -\frac{i}{\sqrt{2}} (\mathbf{k}_0 - \mathbf{k})\mathbf{e}_{sj} \left(\frac{2h}{NM\omega_{sj}} \right)^{1/2} (n_{sj})^{1/2} + \dots \quad (13)$$

where ω_{sj} is the frequency corresponding to the oscillator ξ_{sj} of wave vector \mathbf{q}_s and polarization \mathbf{e}_{sj} .

We consider the process of the emission of n phonons. This corresponds to such matrix elements $F_{\tau\tau_0}$ where of the $3N$ factors in Eq. (10) n have the form $F_{n+1,n}$ and the rest the form $F_{n,n}$. If we number those oscillators which are involved in the transition from 1 to n and write

$$(\mathbf{k}_0 - \mathbf{k}) \mathbf{e}_{sj} = |\mathbf{k}_0 - \mathbf{k}| \cos \alpha_{sj}, \quad r = h^2 (\mathbf{k}_0 - \mathbf{k})^2 / 2M\Theta, \quad (14)$$

where Θ is a unit of energy which can for the sake of convenience be chosen to be of the order of the magnitude of the Debye temperature of the crystal, we get

$$|F_{\tau\tau_0}^{(n)}|^2 = e^{-2W} \left(\frac{2r}{N}\right)^n \sum_{i=1}^n \cos^2 \alpha_i (n_i + 1) \frac{\Theta}{h\omega_i}, \quad (15)$$

where e^{-2W} , the so-called thermal factor, is the product of all $|F_{n_{sj}n_{sj}}|^2$ with n finite and with $N \rightarrow \infty$.

We introduce now the spectrum of the normal vibrations of the crystal, $\varphi(h\omega)$, in such a way that $\varphi(h\omega) d(h\omega)$ is the number of oscillators (sj) with energy $h\omega$ within the interval $d(h\omega)$. The normalization condition is obviously

$$\int_0^\infty \varphi(h\omega) d(h\omega) = 3N. \quad (16)$$

Equation (15) must be multiplied by $\delta(E_\tau + E - E_{\tau_0} - E_0)$ and summed over τ . The summation over τ is equivalent to multiplication by*

$$\prod_{i=1}^n \frac{1}{2} \varphi(h\omega_i) d(h\omega_i)$$

and integration over all $d(h\omega)$ from 0 to ∞ , replacing $\cos^2 \alpha$ by its average value $1/3$ (this is always correct for polycrystalline substances and for a single crystal provided the lattice is cubic). The result obtained must be divided by $n!$ since states for which the excitation of the oscillators differs only in the numbering are identical (in corpuscular language this is the fact that phonons are identical). If the temperature of the crystal is T (in energy units) we must replace $n_i + 1$ by

$$\overline{n_i + 1} = (1 - e^{-h\omega_i/T})^{-1}.$$

Taking all this into account and introducing dimensionless quantities

$$x \equiv h\omega/\Theta; \quad \varepsilon \equiv (E - E_0)\Theta; \quad \varphi_0(x) \equiv (\Theta/3N) \varphi(h\omega), \quad (17)$$

we get

$$\sum_{\tau} |F_{\tau\tau_0}|^2 \delta(E_\tau + E - E_{\tau_0} - E_0) = \frac{e^{-2W}}{\Theta n!} \left(\frac{2r}{N}\right)^n \left(\frac{N}{2}\right)^n \int_0^\infty \dots \int_0^\infty \delta\left(\sum_{i=1}^n x_i - \varepsilon\right) \prod_{i=1}^n \frac{\varphi_0(x_i) dx_i}{(1 - \exp\{-x_i\Theta/T\}) x_i} \equiv \frac{e^{-2W}}{\Theta} z_n, \quad (18)$$

$$z_n = \frac{1}{n!} \int_0^\infty r\lambda(x_1) dx_1 \dots \int_0^\infty r\lambda(x_n) dx_n \delta(x_1 + \dots + x_n - \varepsilon), \quad (19)$$

where

$$\lambda(x) = x^{-1} \varphi_0(x) (1 - e^{-x\Theta/T})^{-1}. \quad (20)$$

A completely analogous expression can be obtained for the absorption of n phonons; only $\overline{n_i + 1}$ must be replaced by $\overline{n_i}$ so that $\lambda(x)$ has the form

$$\lambda(x) = x^{-1} \varphi_0(x) (e^{x\Theta/T} - 1)^{-1}. \quad (21)$$

We introduce the Laplace transform of z_n considered as a function of ε

$$\tilde{z}_n(s) = \int_0^\infty z_n(\varepsilon) e^{-\varepsilon s} d\varepsilon = \frac{r^n}{n!} [\tilde{\lambda}(s)]^n, \quad (22)$$

where

* The factor $1/2$ arises because the summation is over a half-space.

$$\tilde{\lambda}(s) = \int_0^{\infty} \lambda(x) e^{-sx} dx.$$

We sum over n ,

$$\sum_{n=1}^{\infty} \tilde{z}_n(s) = \sum_{n=1}^{\infty} [r\tilde{\lambda}(s)]^n/n! = e^{r\tilde{\lambda}(s)} - 1. \quad (23)$$

Transforming back we get

$$z(\varepsilon) = \sum_{n=1}^{\infty} z_n = \frac{1}{2\pi i} \int_{\sigma-i\infty}^{\sigma+i\infty} \{e^{r\tilde{\lambda}(s)} - 1\} e^{s\varepsilon} ds. \quad (24)$$

Taking Eqs. (3) and (18) into account we find

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k}{k_0} (\bar{a}^2) \left(1 + \frac{m}{M}\right)^2 e^{-2W} \frac{z(\varepsilon, r)}{\Theta}. \quad (25)$$

The thermal factor is calculated without any difficulty

$$W = r \int_0^{\infty} \frac{\varphi_0(x)}{x} \left(\frac{1}{e^{x\Theta/T} - 1} + \frac{1}{2} \right) dx. \quad (26)$$

3. EXAMPLES

We consider now two examples, choosing a definite form of the function $\varphi_0(x)$. The usual Debye spectrum has the form

$$\varphi_0(x) = \begin{cases} 3x^2 & \text{if } x < 1 \\ 0 & \text{if } x > 1 \end{cases} \quad (27)$$

and Θ is called the Debye temperature of the crystal. This function is very inconvenient for computations. We assume a different form for $\varphi_0(x)$, namely,

$$\varphi_0(x) = 1/2 \beta^3 x^2 e^{-\beta x}. \quad (28)$$

This function satisfies the normalization condition (16), goes to zero as x^2 and contains a cutting-off factor $e^{-\beta x}$. The dimensionless quantity β which is of the order of unity can be varied.

In the two limiting cases, discussed above, we can easily pursue the calculations unto the end.

1. Scattering of slow neutrons by hot substances, $T \gg \Theta$. From Eq. (21) we get, accurate up to terms of the order $x\Theta/T$,

$$\lambda(x) \cong \frac{\varphi_0(x)}{x} \frac{T}{x\Theta} e^{-x\Theta/2T} = \frac{T\beta^3}{2\Theta} \exp\left\{-\left(\beta + \frac{\Theta}{2T}\right)x\right\}. \quad (29)$$

Its Laplace transform is

$$\tilde{\lambda}(s) = T\beta^3/2\Theta (\beta + \Theta/2T + s).$$

Moreover, from Eq. (24) we get, omitting the intermediate calculations, the following result

$$z(\varepsilon, r) = \exp\left\{-\left(\beta + \frac{\Theta}{2T}\right)\varepsilon\right\} \left[\frac{T}{\Theta} \frac{\beta^3 r}{2\varepsilon}\right]^{1/2} I_1\left[\left(\frac{T}{\Theta} 2\beta^3 r\varepsilon\right)^{1/2}\right], \quad (30)$$

where I_1 is the Bessel function of imaginary argument.

2. Scattering of neutrons by cold substances, $T \ll \Theta$. We have from Eq. (20) for the emission of an arbitrary number of phonons by the neutron,

$$\lambda(x) \approx \varphi_0(x)/x = 1/2 \beta^3 x e^{-\beta x}. \quad (31)$$

Its Laplace transform is

$$\tilde{\lambda}(s) = 1/2 \beta^3 (\beta + s)^{-2}. \quad (32)$$

Again omitting the calculations we quote the result following from Eq. (24)

$$z(\epsilon, r) = \frac{e^{-\beta\epsilon}}{\epsilon} f\left(\frac{\beta^3}{2} r\epsilon^2\right), \quad (33)$$

where

$$f(x) \equiv \sum_{n=1}^{\infty} x^n/n! (2n-1)! \quad (34)$$

Since the expansion (34) is in powers of r , it is an expansion in the number of phonons involved in the scattering process. The condition that it is necessary to consider only one-phonon processes is, obviously, $x \ll 1$. Since $\frac{1}{2}\beta^3 \approx 1$ we get, if we take into account Eqs. (14) and (17)

$$\frac{(p_0-p)^2}{2M} \frac{1}{\Theta} \frac{(E-E_0)^2}{\Theta^2} \ll 1. \quad (35)$$

Hence we get the following condition for the initial energy

$$(m/M) (E_0/\Theta)^3 \ll 1. \quad (36)$$

If inequality (35) is taken in the opposite sense, the main contribution comes from many-phonon processes. In Eq. (34) there are many terms which are important. Using the Stirling formula and differentiating with respect to n we find the value n_0 for which the expression under the \sum sign is maximum

$$\ln x + \ln(1/n_0) - 2 \ln(2n_0 - 1) + O(1/n_0) = 0, \quad n_0 \approx (x/4)^{1/2}. \quad (37)$$

In this way we find the most probable number of phonons which will be emitted for a given angle of scattering and energy loss (which determine r and ϵ),

$$n_0 = \frac{1}{2} \beta (r\epsilon^2)^{1/2}. \quad (38)$$

If Θ tends to zero we must obtain the scattering by a free nucleus. From Eqs. (14) and (17) it follows that as $\Theta \rightarrow 0$, $r \rightarrow \infty$, $\epsilon \rightarrow \infty$. For large values of x one can obtain the following asymptotic equation for $f(x)$

$$f(x) = (x/4)^{1/2} (3\pi)^{-1/2} \exp\{3(x/4)^{1/2}\}. \quad (39)$$

From Eqs. (26) and (28) we find

$$W = \beta r/4 \quad \text{for } T = 0. \quad (40)$$

If we perform successively the various limiting transitions we get

$$\frac{e^{-2W}}{\Theta} z(r, \epsilon) \rightarrow \frac{1}{\Theta} \delta(r - \epsilon) = \delta\left[\frac{(p_0-p)^2}{2M} - \Delta E\right], \quad (41)$$

$$\frac{d^2\sigma}{d\Omega dE} = \frac{k}{k_0} \bar{a}^2 \left(1 + \frac{m}{M}\right) \delta\left[\frac{(p_0-p)^2}{2M} - \Delta E\right]. \quad (42)$$

This is, as can easily be seen, the cross section for scattering by a free nucleus. Indeed, the δ -function in Eq. (42) assures the conservation of energy provided momentum is conserved (the momentum taken up by the nucleus is equal to $p_0 - p$). Integration over the angles gives

$$\frac{d\sigma}{dE} = \begin{cases} 4\pi (\bar{a}^2)/\Delta E_{\max} & \text{for } 0 < E_0 - E < \Delta E_{\max} \\ 0 & \text{for } E_0 - E > \Delta E_{\max}, \text{ or } E_0 - E < 0, \end{cases} \quad (43)$$

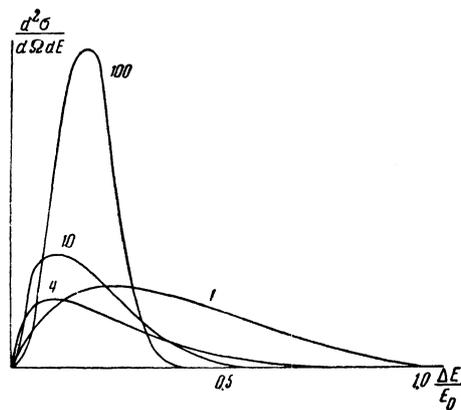
where

$$\Delta E_{\max} = E \cdot 4Mm/(M+m)^2. \quad (44)$$

Integration over the energy gives

$$\sigma = 4\pi (\bar{a}^2). \quad (45)$$

In the figure we have given as an illustration the cross section $d^2\sigma/d\Omega dE$ for a given angle of 90° as a function of the relative energy loss $\Delta E/E_0$ for $M/m = 9$. The value of β was taken to be 3. In this case the thermal factor (40) coincides with the thermal factor for the Debye spectrum. It can be seen from the figure how for increasing E_0/Θ the cross section tends to the δ -function (42) since for the scattering by



Energy distribution of neutrons scattered by a crystal over an angle of 90° . The numbers indicate the value of E/θ . The scale of the curve for $E/\theta = 1$ is increased 10 times with respect to the other curves

a free nucleus the energy loss is simply by the angle of scattering from the laws of conservation of energy and momentum. For $E/\theta = 100$ the width of the curve is still fairly large. In a real crystal the approach to a δ -function will, apparently, occur fast owing to the possibility of knocking a nucleus out of the lattice, a process not considered in our paper.

The calculations, using Eq. (34), can be simplified by using the circumstances that for $x \lesssim 10$ the series (34) converges very rapidly, while for $x \gtrsim 10$ the asymptotic formula

$$\log f(x) = 0.8207 \cdot x^{-1/2} + \frac{1}{6} \log x - 0.5875 - 0.1028 x^{-1/2}. \quad (46)$$

is correct with a large degree of accuracy.

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QUANTUM KINETIC EQUATION FOR PLASMA WITH ACCOUNT OF CORRELATION

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A quantum kinetic equation has been obtained for a system of particles with Coulomb interaction. This equation differs from the known quantum kinetic equation by the fact that correlation of the mutual positions of charged particles has been taken into account.

A quantum kinetic equation for a set of interacting particles can be obtained by solution of the system of equations for the quantum distribution functions f_1 and f_2 .^{1,2} In this case the function f_3 which enters the equation for the distribution function f_2 , is expressed approximately in terms of the functions f_1 and f_2 . For a solution of this system of equations, quantum conditions for the vanishing of correlation at infinity are necessary. However, as was noted by N. N. Bogoliubov, the solution of the equation for the density matrix (or, correspondingly, for the quantum distribution function) can be reduced to a solution of the equation for the quantum function F_S with classical boundary conditions. Here $f_S = \gamma_S F_S$, γ_S is the symmetrization operator for s particles. In the case of systems with central interaction, the equations for F_1 and F_2 have the following form: