Nonlinear correlations of the susceptibility and multiparticle correlation functions

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A general approach is proposed to the determination of the response of an equilibrium system to an electromagnetic field. Diagram expansions of the average current density both in terms of the external field and in terms of the field in the medium are obtained. Graphs that determine the susceptibilities of arbitrary order of nonlinearity are obtained on the basis of the expansions. Relations are derived between the susceptibilities and the kinetic coefficients that determine the response of the system to an external field. These coefficients are expressed in terms of equilibrium correlation functions of the current and of the charge density.

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1. INTRODUCTION. FORMULATION OF PROBLEM

The problem of calculating the kinetic coefficients that determine the response of an equilibrium system to an external action was actively discussed in the literature of the middle fifties. In a classical paper, Kubo[1] obtained expressions for the kinetic coefficient in terms of the corresponding correlation functions of an equilibrium system. These results were used also to calculate the susceptibilities, which determine the response of a system to an electromagnetic field. Kubo himself, as well as most workers subsequently dealing with the problem (for example, Isihara[2]), started from the Hamiltonian of a system of charged particles in an electromagnetic field:

$$\hat{H}_e' = \sum_i \frac{1}{2m_i}\left[\hat{\mathbf{p}}_i - \frac{e_i}{c}\mathbf{A}(\hat{\mathbf{r}}_i, t)\right]^2 + \frac{1}{2}\sum_{i\neq j}\frac{e_ie_j}{|\hat{\mathbf{r}}_i - \hat{\mathbf{r}}_j|} + \sum_i e_i\varphi_{ext}(\hat{\mathbf{r}}_i, t). \qquad (1.1')$$

Here $\mathbf{A}(\mathbf{r}, t)$ is the vector potential of the electromagnetic field in the medium (Coulomb gauge) and $\varphi_{\text{ext}}(\mathbf{r}, t)$ is the scalar potential of the external field.

The standard procedure of calculating the susceptibilities consists in the following. The Hamiltonian of the unperturbed system is chosen in the form

$$\hat{H}' = \sum_i \frac{\hat{\mathbf{p}}_i^2}{2m_i} + \frac{1}{2}\sum_{i\neq j}\frac{e_ie_j}{|\hat{\mathbf{r}}_i - \hat{\mathbf{r}}_j|}. \qquad (1.2')$$

Then, regarding $\hat{H}_e' - \hat{H}'$ as a perturbation and using nonstationary perturbation theory, it is possible to obtain an expansion of the average microscopic current density $\langle\mathbf{j}(\mathbf{r}, t)\rangle$ in powers of the potentials of the field. It is the coefficients of this expansion which are usually interpreted as the corresponding susceptibilities.

Actually the problem is much more complicated, and frequently the described approach leads to incorrect results. The presence in (1.1′) of a term that describes that Coulomb interaction of the particles of the medium leads to the need for considering the longitudinal and transverse fields in the medium in different manners. We consider first the response of an isotropic medium to an external longitudinal field. In this case perturbation theory leads to an expansion of the induced charge density in powers of the external scalar potential $\varphi_{\text{ext}}(\mathbf{r}, t)$. In order to calculate from this expansion the true longitudinal susceptibilities, which determine the response to the true field in the medium, a standard self-consistency procedure is used.[3] Applying perturbation theory for the transverse fields in an isotropic medium, we obtain an expansion of the density of the induced current in powers of the vector potential $\mathbf{A}(\mathbf{r}, t)$. The coefficients of this expansion determine the true transverse susceptibilities of the medium.

Thus, the calculation of the susceptibilities of an isotropic medium is carried out essentially in different manners for longitudinal and transverse fields. However, whereas in an isotropic medium it is possible to consider the longitudinal and transverse fields independently, in the presence of anisotropy this is in general impossible. In an anisotropic medium the transverse field induces currents that have a longitudinal component, and this leads to the appearance of a longitudinal field and vice verse. In addition, even in an isotropic medium the self-consistency referred to above is a complicated problem if a nonlinear response of the medium is considered. The above-mentioned difficulties point to the need of a more consistent examination of the interaction of the medium with the electromagnetic field.

The problem of calculating the linear and nonlinear susceptibilities is considered with the aid of a diagram variant of nonstationary perturbation theory. We shall obtain an expansion of the response of the medium in powers of the external field. The coefficients of this expansion will be called the external susceptibilities, to distinguish them from the susceptibilities that determine the response to the true field in the medium. To obtain the latter, the indicated expansion is rearranged in such a way that it becomes an expansion in terms of the field in the medium. It will be shown that the susceptibilities should be calculated from irreducible polarization parts of the corresponding order of nonlinearity. For the linear susceptibility this result was obtained by Dzyaloshinskiĭ and Pitaevskiĭ.[4]

The question of the connection of the true and external susceptibilities was considered by different authors. The simplest and most natural approach was proposed

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by Silevitch and Golden.[5] They obtained directly from Maxwell's equations the connection between the true and the external susceptibilities that describe effects that are linear and quadratic in the field. However, even this method is too cumbersome to make it possible to propose a systematic approach to the higher-order nonlinear susceptibilities.

The formal procedure developed in Sec. 2 enables us to propose in Sec. 3 a rather simple algorithm for determining the connection between the true and external susceptibilities. It can be derived directly from the connection between the irreducible and reducible polarization parts of the corresponding order of the nonlinearity.

In Sec. 4 we obtain analytic expressions for the external susceptibilities in terms of the equilibrium correlation functions of the currents. The results agree only outwardly with those obtained by Kubo.[1] In contrast to Kubo, we did not assume the field to be a specified function of the coordinates and of the time, and regarded it as one of the dynamic variables, having specified only the extraneous currents that determine the field. As a consequence, the susceptibilities obtained in Sec. 4 differ from those obtained by Kubo.[1] This manifests itself formally in the fact that the susceptibilities indicated above are expressed in terms of equilibrium correlation functions of the current operators in different Heisenberg representations. It seems curious, in our opinion, that the Kubo formula as applied to an isotropic medium describes actually an external longitudinal but a true transverse susceptibility. For an anisotropic medium, one can hardly interpret in reasonable manner the Kubo formula in the form obtained by him.

This paper considers the response of an equilibrium nonrelativistic system of particles to an electromagnetic field. Inasmuch as the system is no longer in equilibrium after application of the external field, we use the diagram technique proposed by Keldysh.[6] We shall not dwell on the necessary formalism, and refer the readers to the cited papers.

The Hamiltonian of a system situated in an external electromagnetic field is of the form

$$\hat{H}_e=\hat{H}-\frac{1}{c}\int \mathbf{j}_e(\mathbf{r},t)\hat{\mathbf{A}}(\mathbf{r})\,d\mathbf{r}+\iint\frac{e\hat{\psi}^+(\mathbf{r})\hat{\psi}(\mathbf{r})\rho_e(\mathbf{r}',t)}{|\mathbf{r}-\mathbf{r}'|}\,d\mathbf{r}\,d\mathbf{r}'. \quad (1.1)$$

Here $\hat{\mathbf{A}}(\mathbf{r})$ is the vector potential of the electromagnetic field. The densities $\mathbf{j}_e(\mathbf{r},t)$ of the extraneous current and $\rho_e(\mathbf{r},t)$ of the charge are assumed to be specified functions of the coordinates and of the time. The first term in (1.1) is the Hamiltonian of the system in the absence of sources of an external field:

$$\hat{H}=\hat{H}_0-\frac{1}{c}\int \hat{\mathbf{j}}_p(\mathbf{r})\hat{\mathbf{A}}(\mathbf{r})\,d\mathbf{r}-\frac{1}{2c}\int \hat{\mathbf{j}}_d(\mathbf{r})\hat{\mathbf{A}}(\mathbf{r})\,d\mathbf{r}$$
$$+\frac{1}{2}\iint\frac{e\hat{\psi}^+(\mathbf{r})\hat{\psi}(\mathbf{r})e\hat{\psi}^+(\mathbf{r}')\hat{\psi}(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|}\,d\mathbf{r}\,d\mathbf{r}', \quad (1.2)$$

where $\hat{H}_0$ is the Hamiltonian of the free particles and of the field, the second and third terms in (1.2) describe the interaction of the particles and the fields, while the last term corresponds to Coulomb interaction of the particles. The current density is broken up into paramagnetic and diamagnetic parts:

$$\hat{\mathbf{j}}(\mathbf{r})=\hat{\mathbf{j}}_p(\mathbf{r})+\hat{\mathbf{j}}_d(\mathbf{r}); \quad (1.3)$$
$$\hat{\mathbf{j}}_p(\mathbf{r})=\frac{i\hbar e}{2m}\left\{\frac{\partial\hat{\psi}^+}{\partial\mathbf{r}}\hat{\psi}(\mathbf{r})-\hat{\psi}^+(\mathbf{r})\frac{\partial\hat{\psi}}{\partial\mathbf{r}}\right\},$$
$$\hat{\mathbf{j}}_d(\mathbf{r})=-\frac{e^2}{mc}\hat{\psi}^+(\mathbf{r})\hat{\psi}(\mathbf{r})\hat{\mathbf{A}}(\mathbf{r}).$$

We assume for simplicity that the system consists of one sort of spinless particles. The corresponding generalizations entail no particular difficulty. Nor is it difficult to generalize the results to the case when other types of interactions are present, such as electron-phonon interaction.

As seen from (1.2) the interaction of the particles with the field contains paramagnetic and diamagnetic terms. We shall accordingly distinguish on the diagrams the paramagnetic and diamagnetic vertices. Besides the customary factor $1/i\hbar$ of the Feynman technique, an operator—$\mathbf{j}_p(x)/c$ and one field operator $\hat{\mathbf{A}}(x)$ correspond to the paramagnetic vertex, while the operator $e^2\hat{\psi}^+(x)\hat{\psi}(x)/mc^2$ and two field operators correspond to a diamagnetic vertex. We add to each diamagnetic vertex a factor of 2 to account for the possible permutation that occurs when field operators corresponding to one diamagnetic vertex are paired. It must be recognized here that the diagrams may contain substructures in which both field operators of a certain diamagnetic vertex become paired with identical expressions. Since the permutation of the field operator does not lead in this case to a new pairing system, a factor $\frac{1}{2}$ must be assigned to each such substructure.

2. AVERAGE FIELD. AVERAGE CURRENT DENSITY

We proceed to find the diagram expansions for the average field and the average current density:

$$\langle A^\alpha(x)\rangle_e=\mathrm{Tr}\,\{\hat{\rho}(0)\hat{A}_e^\alpha(x)\}, \quad (2.1)$$
$$\left\langle-\frac{1}{c}j^\alpha(x)\right\rangle_e=-\frac{1}{c}\mathrm{Tr}\{\hat{\rho}\,(0)\hat{j}_e^\alpha(x)\}. \quad (2.2)$$

We use here a Coulomb gauge for the potentials, and consequently, we should use a four-dimensional scheme for the complete description of the field. By the time-dependent ($\alpha=0$) components of the current density and of the potential we mean respectively the charge density and the scalar potential:

$$-\frac{1}{c}\hat{j}^0(x)=\frac{1}{c}\hat{j}_0(x)=-e\hat{\psi}^+(x)\hat{\psi}(x), \quad (2.3)$$
$$\hat{A}_e^0(x)=\int\frac{\delta(t-t')}{|r-r'|}\{\rho_e(x')+e\hat{\psi}_e^+(x')\hat{\psi}_e(x')\}\,dx', \quad (2.4)$$

and by the spatial components ($\alpha=1,2,3$) we mean the current density and the vector potential. As usual, $\mathbf{x}=\mathbf{r}$ and $x^0=-x_0=ct$. The dynamic variables averaged in (2.1) and (2.2) are taken in the Heisenberg representation with Hamiltonian (1.1), a fact designated by the subscript e. The averaging is carried out with the aid of a statistical operator taken at the instant of time $t=0$:

$$\hat{\rho}(0)=\mathcal{S}(0,-\infty)\hat{\rho}_0 S^+(0,-\infty). \qquad (2.5)$$

Here $\hat{\rho}_0$ is the statistical operator at $t=-\infty$, when all the interactions in the system are assumed to be turned off, and the extraneous current and charges are assumed to be zero.

Substituting (2.5) in (2.1) and (2.2) and expanding the S matrix in powers of the interaction parameter, [6,8] we obtain the expansion of the average field in the medium and the average current density in the form of a sum of all possible pairing systems, each of which is set in correspondence with a definite diagram. Classifying the diagrams in accordance with the number of interactions with the source of the external field, we obtain as a result the diagram series shown in Figs. 1a and 1b. The wavy lines are the free-field lines. Each of them is set in correspondence to a Green's function $i\hbar D_0^{\alpha,\beta}(xi, x'i')$, which is defined in the Appendix A. The crosses in the figure denote the sources of the external field $-j_e^\alpha(x)/c$. At $\alpha=1$, 2, and 3 the series shown in Figs. 1a and 1b determine the average vector potential and the average current density. At $\alpha=0$ they correspond to the average scalar potential and the average charge density. In all the internal vertices, integration is carried out over $t\in(-\infty,\infty)$ and $\mathbf{r}\in V$ (the volume of the system) and summation is carried out over the tensor indices $\alpha=0,1,2,3$. In addition, the time coordinate of each of the vertices can lie both on the positive and on the negative part of the integration contour C, which runs from $t=-\infty$ to $t=+\infty$ and back from $t=+\infty$ to $t=-\infty$. Accordingly, each vertex of the diagram is set in correspondence to a factor σ_{ii} (σ is the third Pauli matrix). Summation is carried out over the indices i of the internal vertices ($i=1,2$). The shaded parts on Figs. 1a and 1b denote the reducible polarization parts of the first, second, etc., orders. The polarization part of the n-th order is defined as the aggregate of all possible topologically different diagrams that have n inputs and one output vertex of the electromagnetic interaction.

The first term of the diagram series in Fig. 1a corresponds to an external field

$$A_e(x)=-\frac{1}{c}D_0(xi,\xi j)\sigma_{jj}j_e(\xi). \qquad (2.6)$$

On the other hand, the diagram series presented in Fig. 1b corresponds to expansion of the microscopic current density in powers of the external-field potential:

$$-\frac{\sigma_{ii}}{c}\langle j(x)\rangle_e=\sum_{n=1}^{\infty}P^{red}_{ij_1\ldots j_n}(x,\xi_1,\ldots,\xi_n)\prod_{s=1}^{n}A_e(\xi_s). \qquad (2.7)$$

In (2.6) and (2.7) we have omitted the signs of summation over $j=1$ and 2 and of integration with respect to ξ. In addition, we have left out the tensor indices α. Contraction is carried out over the tensor indices of the internal vertices. $P^{\rm red}_{ij_1\ldots j_n}(x,x_1,\ldots,x_n)$ denotes the polarization part of n-th order.

Both expressions can be rewritten in terms of retarded functions. Using (A.4) and introducing the kinetic coefficients

$$\chi_e(x,x_1,\ldots,x_n)=S_{1\ldots n}\sigma_{ii}\sum_{j_1\ldots j_n=1}^{2}P^{red}_{ij_1\ldots j_n}(x,x_1,\ldots,x_n), \qquad (2.8)$$

we rewrite (2.6) and (2.7) in the form

$$A_e(x)=-\frac{1}{c}D_0^R(x,\xi)j_e(\xi), \qquad (2.9)$$

$$\left\langle -\frac{1}{c}j(x)\right\rangle=\sum_{n=1}^{\infty}\chi_e(x,\xi_1,\ldots,\xi_n)\prod_{s=1}^{n}A_e(\xi_s). \qquad (2.10)$$

In (2.8), symmetrization is carried out over the argument $x_1\cdots x_n$ of the expression in the right-hand side, which follows the symbol S. Expression (2.8) does not depend on the index i and has a retarded character, i.e., it vanishes if at least one of the temporal arguments $t_1\cdots t_n$ exceeds t. Moreover, the contribution of any diagram to (2.8) has a retarded character. A proof of these statements is given in Appendix B.

The expansion (2.10) determines the response of the system to an external field. In order to obtain the response to an external field, it is necessary to reconstruct the diagram series in such a way that they contain only the average field. Any diagram on Fig. 1a begins with a free-field line, which leads to the following:

1) either to an extraneous source of the field—this is the diagram for the external field $A_e(x)$;

2) or to an outgoing vertex of the linear polarization part, the incoming vertex of which is connected with the source of the external field;

3) or to the outgoing vertex of a linear polarization part, followed by a free field line that leads to the outgoing vertex of the irreducible polarization part of n-th order ($n\geq 2$);

4) or else to an outgoing vertex of the irreducible polarization part of n-th order ($n\geq 2$).

Adding the diagrams of the first two types, we obtain the line of the total Green's function (thick line) which goes to the extraneous field source. The obtained term describes the average field in the linear approximation. The summation of all possible diagrams of the succeeding two types, having one and the same irreducible polarization part, leads to a diagram in which the line of

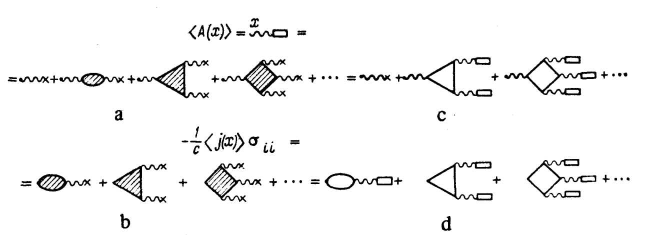

FIG. 1. Diagram expansion for the average field and the average current density: a, b—in terms of the external field; c, d—in terms of the average field.

the complete Green's function is joined to the outgoing vertex of the irreducible polarization part ($n \geq 2$).

Certain substructures which are not related to one another are joined to the incoming vertices of the irreducible polarization part with the aid of the free-field lines. By summing all the possible diagrams having one and the same irreducible polarization part, we find that average-field blocks are joined to the incoming vertices of the irreducible polarization part (Fig. 1c). The series for the average current density is realigned in similar fashion (Fig. 1d). The unshaded blocks denote the irreducible polarization parts. The irreducible polarization part cannot be broken up into polarization parts of the same order or of lower order by removing one field line. The diagram expansion shown in Fig. 1d solves the problem of finding the response to the average field:

$$-\frac{1}{c}\langle j(x)\rangle_e=\sum_{n=1}^{\infty}\chi(x,\xi_1,\ldots,\xi_n)\prod_{s=1}^{n}\langle A(\xi_s)\rangle_e. \tag{2.11}$$

We have introduced here the kinetic coefficients

$$\chi(x,x_1,\ldots,x_n)=S_{1\ldots n}\sigma_{ii}\sum_{j_1\ldots j_n=1,2}P_{ij_1\ldots j_n}(x,x_1,\ldots,x_n), \tag{2.12}$$

which are determined in terms of the irreducible polarization parts and which can be naturally called the "true" susceptibilities in contrast to the external susceptibilities (2.8). The contribution of any diagram to (2.12) has a retarded character. In the calculation of (2.8) and (2.12) it is necessary to take into account only connected diagrams.

In the foregoing construction of the diagram expansions for (2.1) and (2.2), we described the field with the aid of potentials in a Coulomb gauge. This was dictated by the need of applying the Wick theorem and to our desire to separate the Coulomb interaction. In addition, in the nonrelativistic theory, the Coulomb gauge is distinguished by the fact that the field dynamic variables describe only the transverse part of the field.[7] The transverse part of the field, on the other hand, is described by a vector potential in the Coulomb gauge.

A disadvantage in the description of the field in a Coulomb gauge is the need for considering, besides the vector potential, also the scalar potential that describes the longitudinal part of the field. This forces us to use a scheme in which the tensor indices corresponding to all the vertices run through four values, whereas only three tensor indices are assigned to the polarizability in the usual sense of the word, in correspondence with the projections of the electric-field vector. It makes sense therefore to change over in (2.9)–(2.11) to a gauge with a zero scalar potential. In this case, the connection between the potential and the electric field is simplest in form:

$$\mathbf{E}=-\frac{1}{c}\frac{\partial}{\partial t}\mathbf{A}_{\varphi=0}.$$

As a result the Green's functions (A.14) of the potential in this case coincide essentially with the Green's functions of the electric field, and the expansions obtained above for the response of the system are in fact expansions in the electric field. The absence of a scalar potential makes it possible to use an ordinary three-dimensional schemen for the description of the total field. The changes that must be carried out in formulas (2.9)–(2.11) consist of replacing the Green's function $D^{R\alpha\beta}(x, x')$ ($\alpha, \beta=0, 1, 2, 3$) by the Green's function $G^{R\alpha\beta}(x, x')$ ($\alpha, \beta=1, 2, 3$). We note that for the field lines that join the vertices of one and the same irreducible polarization part, the Coulomb gauge is preserved, making it possible to separate the Coulomb interaction of the particles.

To be able to carry out the indicated change of gauge, it suffices that the four-dimensional divergence of the susceptibility with respect to the coordinates of each of the external vertices vanish:

$$\sum_{\alpha=0}^{3}\frac{\partial\chi^{\alpha\alpha_1\ldots\alpha_n}}{\partial x^{\alpha}}(x,x_1,\ldots,x_n)=\ldots=\sum_{\alpha_l=0}^{3}\frac{\partial\chi^{\alpha\alpha_1\ldots\alpha_n}}{\partial x^{\alpha_l}}(x,x_1,\ldots,x_n)=\ldots=0. \tag{2.13}$$

This equation is equivalent to the charge-conservation law and ensures gauge invariance of the response.

It can be verified directly that the contributions of the individual diagrams to the susceptibility do not satisfy such a condition. We shall present below a rule that makes it possible to select groups of diagrams whose combined contribution to the susceptibility satisfies (2.13). Assume that we have a certain diagram for an irreducible polarization part of n-th order, having an external vertex x (incoming or outgoing) or the paramagnetic type. We add to this diagram n-th order diagrams for the polarization parts, which also have internal and external vertices, but which have a different topological structure. To the resultant aggregate we now add diagrams obtained by replacing each of the diagrams of the indicated aggregate the vertex x and one of the remaining paramagnetic vertices by a diamagnetic vertex. We can then show that the contribution of the group of diagrams constructed in this manner to the susceptibility has a zero four-dimensional divergence with respect to the argument x. Thus, to obtain a zero four-dimensional divergence with respect to the argument x' it is necessary to add the diagrams of Figs. 2b, 2c, and 2d to the diagram of Fig. 2a. After adding the diagrams of Figs. 2e, 2f, and 2g, the contribution of the obtained group of diagrams to the susceptibility will have a zero four-dimensional divergence also with respect to the argument x.

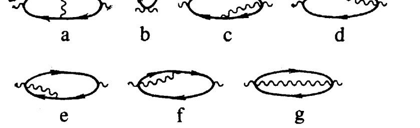

FIG. 2. Example of group of diagrams whose contribution to the susceptibility has zero 4-divergences with respect to the coordinates of the external vertices.

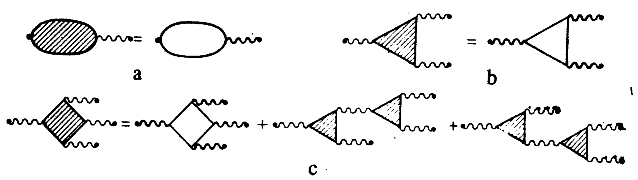

FIG. 3. Connection of reducible and irreducible polarization parts: a—linear, b—quadratic, c—cubic.

3. CONNECTION BETWEEN THE SUSCEPTIBILITIES

Let us find the connection between the susceptibility $\chi(x, x_1, \ldots, x_n)$ and the external susceptibility $\chi_e(x, x_1, \ldots, x_n)$. This connection can be obtained by considering the relation between the reducible and irreducible polarization parts of n-th order. The connection of the linear polarization parts is shown in Fig. 3a. The linear susceptibilities are correspondingly connected by the relation

$$\chi_e(x, \xi) G_0^R(\xi, x') = \chi(x, \xi) G^R(\xi, x'). \tag{3.1}$$

The relation between the quadratic reducible and irreducible polarization parts is shown in Fig. 3b. Expressing this relation with the aid of (A.4), (2.8), and (2.12) in terms of the retarded Green's functions and the quadratic susceptibility, we obtain in the Coulomb gauge

$$D_0^R(x,\xi)\chi_e(\xi,\xi_1,\xi_2)\prod_{s=1,2} D_0^R(\xi_s, x_s) = D^R(x,\xi)\chi(\xi,\xi_1,\xi_2)\prod_{s=1,2} D^R(\xi_s, x_s). \tag{3.2}$$

We now change over with the aid of (A.14) and (2.13) to Green's functions in a gauge with zero potential, and obtain

$$G_0^R(x,\xi)\chi(\xi,\xi_1,\xi_2)\prod_{s=1,2} G_0^R(\xi_s,x_s)$$
$$= G^R(x,\xi)\chi(\xi,\xi_1,\xi_2)\prod_{s=1,2} G^R(\xi_s,x_s). \tag{3.3}$$

Starting with the third order, the connection between the susceptibilities χ and χ_e becomes more complicated. This is due to the possibility of constructing higher-order polarization parts ($n \geq 3$) out of the lower-order parts. However, on the basis of the results of Appendix B, we can state that we can always change from relations that connect reducible and irreducible polarization parts to relations containing susceptibilities and retarded Green's functions. In the latter relations, in turn, we can always change to Green's functions in a gauge $\varphi = 0$.

Consider the cubic susceptibility. The connection between the irreducible and reducible polarization parts is shown in Fig. 3c.

Comparing, as above, the analytic expression, we obtain

$$G_0^R(x,\xi)\chi_e(\xi,\xi_1,\xi_2,\xi_3)\prod_{s=1}^{3} G_0^R(\xi_s,x_s) - G^R(x,\xi)\chi(\xi,\xi_1,\xi_2,\xi_3)\prod_{s=1}^{3} G^R(\xi_s,x_s)$$
$$= 2G^R(x,\xi) S_{123}\{\chi(\xi,\xi_1,\eta')G^R(\eta',\eta'')\chi(\eta'',\xi_2,\xi_3)\}\prod_{s=1}^{3} G^R(\xi_s,x_s). \tag{3.4}$$

Relations (3.1), (3.3), and (3.4) solve the problem of finding the connection between the susceptibilities that determine the response to the average and external fields (at $n = 1, 2, 3$). To obtain an explicit expression for χ in terms of χ_e it suffices to apply the operator

$$-\frac{1}{4\pi}\left(\mathrm{rot\,rot} + \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right) - \hat{\chi}$$

to each of the external coordinates in both parts of (3.1), (3.3), and (3.4) and use Eq. (A.19) for the retarded Green's function $G^R(x, x')$. We use $\hat{\chi}$ to denote an integral operator with a kernel equal to the linear susceptibility $\chi(x, \xi)$.

4. EXTERNAL SUSCEPTIBILITIES

We proceed to construct analytic expressions for the external susceptibilities. We consider the linear and quadratic susceptibilities. Figure 4a shows the structure of the linear reducible polarization part $P_{ii'}^{\mathrm{red}}(x, x')$. The indices j of the vertices of the first term denote that it contains all possible diagrams having normal (or j-type) incoming and outgoing vertices, i.e., either paramagnetic or diamagnetic, but with one of the field ends paired inside the polarization part. The index u of the second term means that it contains diagrams with anomalous, i.e., diamagnetic external vertex, the field ends of which are not paired inside the polarization part. The linear irreducible polarization part has an analogous structure, and incorporates the entire second term of Fig. 4a.

Thus, the first term in the right-hand side of Fig. 4a is the aggregate of all possible diagrams (which do not contain extraneous field sources), whose vertices correspond to the operators $-\hat{j}(x, i)/c$ and $-\hat{j}(x', i')/c$. Exactly the same aggregate of diagrams is encountered in the expansion of the equilibrium correlation function in powers of the interaction parameter

$$\mathrm{Tr}\left\{\hat{\rho}T_C\left\{-\frac{1}{c}\hat{j}_H(x,i), -\frac{1}{c}\hat{j}_H(x',i')\right\}\right\}$$
$$= \left\langle T_C\left\{-\frac{1}{c}\hat{j}(x,i), -\frac{1}{c}\hat{j}(x',i')\right\}\right\rangle.$$

Here T_C denotes the chronological ordering operation, referred to in Sec. 2, along the contour C. This opera-

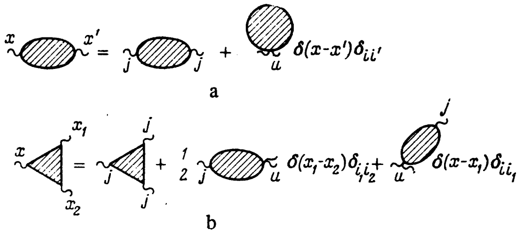

FIG. 4. Structure of polarization parts: a—linear, b—quadratic.

tion coincides with the usual operation of chronological ordering if x^0 and x'^0 lie on the positive part of the contour, and with the operation of anti-ordering if they lie on the negative part. The points of the positive part of the contour C are assumed to precede those on the negative part. The subscripts H of the averaged operators means that they are taken in the Heisenberg representation with the Hamiltonian (1.2), and $\hat{\rho}$ is the equilibrium statistical operator corresponding to this Hamiltonian. We do not write out explicitly the tensor indices α, assuming them to be included in the corresponding argument x. Since all the dynamic-variable operators employed below are taken in the Heisenberg representation, we shall henceforth omit the subscript H.

Considering, as above, the second term of Fig. 4a, we obtain an analytic expression for the linear polarization part:

$$P^{red}_{ii'}(x,x')=\frac{\sigma_{ii}\sigma_{i'i'}}{i\hbar c^2}\langle T_C\{\hat{j}(x,i),\hat{j}(x',i')\}\rangle+\frac{e^2 n(x)}{mc^2}\sigma_{ii}\delta_{ii'}\delta(x-x'). \tag{4.1}$$

Here $n(x)$ is the density of the number of particles in the equilibrium system. Substituting (4.1) in (2.8), we obtain the external linear susceptibility

$$\chi_e(x,x')=\frac{e^2 n(\mathbf{r})}{mc^2}\delta(x-x')+\frac{\theta(t-t')}{i\hbar c^2}\langle[\hat{j}(x),\hat{j}(x')]\rangle. \tag{4.2}$$

On changing over from (4.1) to (4.2) we use the following easily proved statement. For any operator $\xi(t)$, $\xi_1(t_1),\ldots,\hat{\xi}_n(t_n)$ of the Bose type we have

$$\sum_{i_1\ldots i_n=1}^{2} T_C\{\hat{\xi}(t,i),\hat{\xi}_1(t_1,i_1),\ldots,\hat{\xi}_n(t_n,i_n)\}\prod_{s=1}^{n}\sigma_{i_s i_s}$$
$$=\sum_{k_1\ldots k_n=1}^{n}\theta(t-t_{k_1})\ldots\theta(t_{k_{n-1}}-t_{k_n})[\ldots[[\hat{\xi},\hat{\xi}_{k_1}],\hat{\xi}_{k_2}],\ldots,\hat{\xi}_{k_n}]. \tag{4.3}$$

As usual, $\theta(t)$ stands for the function

$$\theta(t)=\begin{cases}1, & t>0,\\ 0, & t<0.\end{cases}$$

We consider now the quadratic susceptibility. The structure of the diagrams for the quadratic polarization part $P^{red}_{ii_1i_2}(x,x_1,x_2)$ is shown in Fig. 4b. Reasoning as in the derivation of (4.1), we get

$$S_{12}P^{red}_{ii_1i_2}(x,x_1,x_2)=\sigma_{ii}S_{12}\Big\{\frac{1}{(i\hbar)^2}\Big(-\frac{1}{c}\Big)^3\langle T_C\{\hat{j}(x,i),\hat{j}(x_1,i_1),\hat{j}(x_2,i_2)\}\rangle$$
$$-\frac{1}{2i\hbar c}\Big\langle T_C\Big\{\hat{j}(x,i),\frac{e^2}{mc^2}\hat{\psi}^+\hat{\psi}(x_1,i_1)\Big\}\Big\rangle\sigma_{i_1i_1}\delta_{i_1i_2}\delta(x_1-x_2)$$
$$-\frac{1}{i\hbar c}\Big\langle T_C\Big\{\frac{e^2}{mc^2}\hat{\psi}^+\hat{\psi}(x,i),\hat{j}(x_2,i_2)\Big\}\Big\rangle\delta_{ii_1}\sigma_{i_2i_2}\delta(x-x_1)\Big\}. \tag{4.4}$$

This yields the quadratic external susceptibility

$$\chi_e(x,x_1,x_2)=S_{12}\Big\langle\frac{\theta(t-t_1)\theta(t_1-t_2)}{(i\hbar)^2}\Big[\Big[-\frac{1}{c}\hat{j}(x),-\frac{1}{c}\hat{j}(x_1)\Big],-\frac{1}{c}\hat{j}(x_2)\Big]$$
$$+\frac{1}{2i\hbar}\theta(t-t_1)\delta(x_1-x_2)\Big[-\frac{1}{c}\hat{j}(x),\frac{e^2}{mc^2}\hat{\psi}^+\hat{\psi}(x_1)\Big]$$
$$+\frac{1}{i\hbar}\theta(t-t_1)\delta(x-x_2)\Big[\frac{e^2}{mc^2}\hat{\psi}^+\hat{\psi}(x),-\frac{1}{c}\hat{j}(x_1)\Big]\Big\rangle. \tag{4.5}$$

In analogy with the procedure used above for the linear and quadratic susceptibilities, we can consider also the higher-order external susceptibilities. The resultant analytic expressions are too cumbersome to present here, all the more since they coincide outwardly with those obtained by Kubo.[1] We note, however, the following. In his paper[1] he considered the problem of calculating the dynamic coefficients that determine the response of an equilibrium system to an external action. As applied to an electromagnetic field, the expression obtained for the linear susceptibility agrees outwardly with (4.2) and is known as the Kubo formula. In the Kubo formula, however, the commutator of the current-density operators is averaged in a Heisenberg representation defined by the Hamiltonian (1.2′), and not by the Hamiltonian (1.2) as is the case in (4.2). To compare the external linear susceptibility obtained above with Kubo's result, let us examine the diagrams corresponding to the Kubo susceptibility. The Hamiltonian (1.2′) does not contain terms corresponding to the interaction of the particles of the medium with the photons. This means that the Kubo susceptibility corresponds to a reducible polarization part that does not contain transverse (photon) lines.

Consider an isotropic medium. In such a medium the longitudinal projection of the diagram, which determines the density of the current when an external transverse field is applied, is equal to zero, just as the transverse projection of the diagram that determines the response to the longitudinal field. It follows therefore that the longitudinal projection (4.2) coincides with the longitudinal projection of the Kubo formula, which thus determines the external longitudinal susceptibility. The same reasoning makes it possible to establish that for the case of a transverse field in an isotropic medium the Kubo formula determines the true transverse susceptibility. As applied to an anisotropic medium, the Kubo formula does not describe either the true or the external susceptibility, and can hardly be interpreted in reasonable fashion.

In conclusion, I am indebted to L. V. Keldysh for suggesting the problem and constant interest in the work. I am also grateful to I. E. Dzyaloshinskiĭ for a useful discussion of the work.

APPENDIX A: GREEN'S FUNCTIONS

The Green's functions in the absence of external field sources are defined in the usual[6] manner

$$i\hbar D^{\alpha\beta}(xi,x'j)=\mathrm{Tr}\{\hat{\rho}T_C\{\hat{A}_H{}^{\alpha}(x,i),\ \hat{A}_H{}^{\beta}(x',j)\}\}. \tag{A.1}$$

The tensor indices $\alpha,\beta=1,2,3$ correspond here to the three projections on the spatial coordinate axis. The Green's functions (A.1) with different i and j are connected by the relations

$$D(x1,x'1)=\theta(t-t')D(x2,x'1)+\theta(t'-t)D(x1,x'2),$$
$$D(x2,x'2)=\theta(t-t')D(x1,x'2)+\theta(t'-t)D(x2,x'1). \tag{A.2}$$

The retarded Green's function

$$i\hbar D^{R\alpha\beta}(x, x') = \theta(t-t')\mathrm{Tr}\{\hat\rho[\hat A_H^\alpha(x), \hat A_H^\beta(x')]\}\qquad (A.3)$$

is connected with (A.1) by the relation

$$D^R(x, x') = \sum_{j=1}^{2} D(xi, x'j)\sigma_{ij}.\qquad (A.4)$$

Relations (A.1) and (A.3) determine the Green's functions in the Coulomb gauge

$$\mathrm{div}\,\hat{\mathbf{A}}(\mathbf{r}) = 0,\qquad (A.5)$$

which we have used to construct the diagram expansions in Sec. 2. The vector-potential operator in this gauge is

$$\hat{\mathbf{A}}(\mathbf{r}) = \sum_{\mathbf{q},\lambda} c\left(2\pi\frac{\hbar}{\omega_{\mathbf{q}}}\right)^{1/2}\left\{\hat b_\lambda(\mathbf{q})\frac{e^{i\mathbf{q}\mathbf{r}}}{V^{1/2}} + \hat b_\lambda^+(\mathbf{q})\frac{e^{-i\mathbf{q}\mathbf{r}}}{V^{1/2}}\right\}\mathbf{n}_\lambda(\mathbf{q}).\qquad (A.6)$$

Here $\hat b_\lambda(\mathbf{q})$ and $\hat b_\lambda^+(\mathbf{q})$ are the operators of the annihilation and production of photons in a state with a wave vector q and polarization $\mathbf{n}_\lambda(\mathbf{q})$, $\lambda = 1, 2$. The free Green's functions are defined as the mean values of the free field operators with the aid of relations analogous to (A.1) and (A.3). The averaging is carried out with the aid of the statistical operator $\hat\rho_0$.

Using (A.6) we easily verify that the free retarded Green's function satisfies the equation ($\alpha, \beta = 1, 2, 3$)

$$\frac{1}{4\pi}\left(\Delta - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)D_0^{R\alpha\beta}(x, x') = \delta(t-t')\delta_\perp^{\alpha\beta}(\mathbf{r}-\mathbf{r}'),\qquad (A.7)$$

where we have introduced the function

$$\delta_\perp^{\alpha\beta}(\mathbf{r}-\mathbf{r}') = \delta^{\alpha\beta}\delta(\mathbf{r}-\mathbf{r}') - \frac{1}{4\pi}\frac{\partial^2}{\partial x^\alpha\,\partial x'^\beta}\frac{1}{|\mathbf{r}-\mathbf{r}'|},$$

which, upon integration with respect to $\mathbf{r}'$ and contraction with respect to β with any vector function, separates the transverse of this function.

The Green's functions referred to above form a 3×3 matrix in the tensor indices α and β. It makes it possible to describe only the transverse part of the field. For a complete description of the field in the Coulomb gauge it is necessary to supplement this matrix to form a 4×4 ($\alpha = 1, 2, 3$) matrix:

$$D_0^{0\alpha}(xi, x'j) = D_0^{\alpha 0}(xi, x'j) = 0,\qquad (A.8)$$

$$D_0^{00}(xi, x'j) = \frac{\delta(t-t')}{|\mathbf{r}-\mathbf{r}'|}\sigma_{ij},$$

$$D_0^{R0\alpha}(x, x') = D_0^{R\alpha 0}(x, x') = 0,\qquad (A.9)$$

$$D_0^{R00}(x, x') = \frac{\delta(t-t')}{|\mathbf{r}-\mathbf{r}'|}.$$

The complete Green's functions (A.1) satisfy the Dyson equation

$$D^{\alpha\beta}(xi, x'i') = D_0^{\alpha\beta}(xi, x'i') + D_0^{\alpha\gamma}(xi, \xi j)P_{\gamma\gamma'}^{red}(\xi j, \xi' j')D_0^{\gamma'\beta}(\xi' j', x'i').\qquad (A.10)$$

$$P_{\alpha\gamma}^{red}(xi, \xi j)D_0^{\gamma\beta}(\xi j, x'i') = P_{\alpha\gamma}(xi, \xi j)D^{\gamma\beta}(\xi j, x'i').\qquad (A.11)$$

Summation is carried out over repeated indices and integration over the continuous variables. At $\alpha, \beta = 1, 2, 3$, Eq. (A.10) is the usual Dyson equation. At α or β equal to zero, Eq. (A.10) should be regarded as a definition of the complete Green's function in the Coulomb gauge. At $\alpha = \beta = 0$, Eq. (A.10) goes over into the equation for the effective interaction potential.[8] Multiplying (A.10) and (A.11) from the right by σ and using (A.4), we obtain the Dyson equation for the retarded Green's function:

$$D^{R\alpha\beta}(x, x') = D_0^{R\alpha\beta}(x, x') + D_0^{R\alpha\gamma}(x, \xi)\chi_{\gamma\gamma'}^{(e)}(\xi, \xi')D_0^{R\gamma'\beta}(\xi', x'),\qquad (A.12)$$

$$\chi_{\alpha\gamma}^{(e)}(x, \xi)D_0^{R\gamma\beta}(\xi, x') = \chi_{\alpha\gamma}(x, \xi)D^{R\gamma\beta}(\xi, x').\qquad (A.13)$$

We now introduce the Green's functions in a gauge with zero scalar potential, which we define in the following manner:

$$G^{R\alpha\beta}(x, x') = D^{R\alpha\beta}(x, x') + \frac{\partial}{\partial x_\alpha}\int_{x'^0}^{x^0} dx^0 D^{R0\beta}(x, x')$$
$$- \frac{\partial}{\partial x_\beta'}\int_{x'^0}^{x^0} dx'^0 D^{R\alpha 0}(x, x') - \frac{\partial^2}{\partial x_\alpha\,\partial x_\beta'}\int_{x'^0}^{x^0} dx^0\int_{x'^0}^{x^0} dx'^0 D^{R00}(x, x').\qquad (A.14)$$

In the last term the integration is consecutive. It is easy to verify that

$$G^{R\alpha 0} = G^{R0\alpha} = G^{R00} = 0.\qquad (A.15)$$

We determine analogously also the free energy, for which the second and third terms drop out by virtue of (A.9). The Green's function (A.14) satisfies the Dyson equation which follows from (A.12) and (2.13):

$$G^{R\alpha\beta}(x, x') = G_0^{R\alpha\beta}(x, x') + G_0^{R\alpha\gamma}(x, \xi)\chi_e^{\gamma\gamma'}(\xi, \xi')G_0^{R\gamma'\beta}(\xi', x'),\qquad (A.16)$$

$$\chi_e^{\alpha\gamma}(x, \xi)G^{R\gamma\beta}(\xi, x') = \chi^{\alpha\gamma}(x, \xi)G^{R\gamma\beta}(\xi, x').\qquad (A.17)$$

The free Green's function in the gauge $\varphi = 0$ satisfies the differential equation

$$-\frac{1}{4\pi}\left[(\mathrm{rot\,rot})_{\alpha\gamma} + \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\delta_{\alpha\gamma}\right]G_0^{R\gamma\beta}(x, x') = \delta_{\alpha\beta}\delta(x-x').\qquad (A.18)$$

The complete Green's function in a gauge with zero scalar potential satisfies the equation

$$-\frac{1}{4\pi}\left[(\mathrm{rot\,rot})_{\alpha\gamma} + \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\delta_{\alpha\gamma}\right]G^{R\gamma\beta}(x, x')$$
$$-\int \chi_{\alpha\gamma}(x, \xi)G^{R\gamma\beta}(\xi, x')d\xi = \delta_{\alpha\beta}\delta(x-x').\qquad (A.19)$$

APPENDIX B: RETARDED CHARACTER OF THE SUSCEPTIBILITY

We consider here the "causal" diagram properties that are typical of the Keldysh technique.[6] As will be shown below, the retarded character of the susceptibilities (2.8) and (2.12) follows directly from the method used for their construction, and the Green's functions of the particles are determined in analogy with the field Green's functions (A.1), and consequently also satisfy a relation similar to (A.2). At $t > t'$ it follows from it that the Green's functions corresponding to the lines connecting the vertex having the largest time-dependent argument from among all the vertices, with the vertices that lie to the left on the time axis, do not depend on whether the outermost right-hand vertex lies on the positive $C_+(-\infty, +\infty)$ or negative $C_-(+\infty, -\infty)$ part of the integration contour C. Since all the internal vertices of the diagram are taken both on C_+ and on C_-, and the vertex lying on C_- is assigned the factor -1, the contribution of the diagram can be different from zero only if the extreme right vertex on the time axis is an external vertex (over the index i of which no summation is carried out). If we denote by $P_{ii_1\ldots i_n}(x, x_1, \ldots, x_n)$ the contribution of the diagram with external vertices x, $x_1, \ldots, x_n$, then at $t > \max\{t_1, \ldots, t_n\}$ we have

$$P_{1i_1\ldots i_n}(x, x_1, \ldots, x_n) = -P_{2i_1\ldots i_n}(x, x_1, \ldots, x_n).$$

It follows from this, in particular, that expressions of the type (2.8) and (2.12) are independent of the index i.

Next,

$$\sum_{i_1\ldots i_n=1,2} P_{ii_1\ldots i_n}(x, x_1, \ldots, x_n) = 0,$$

if at least one of the time-dependent arguments $t_1, \ldots, t_n$ exceeds t. From this follows the retarded character of the contribution of any diagram to the kinetic coefficients defined by relations of the type (2.8) and (2.12).

It follows also from the foregoing that the so-called vacuum loops are absent in the Keldysh technique.[6] In fact, the vacuum loop is not connected with any of the external vertices. Consequently, it corresponds to a zero factor. In the same manner it is easy to verify that only connected diagrams contribute to the kinetic coefficients (2.8) and (2.12).

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Establishment of equilibrium between the nuclear and electron subsystems on dynamic cooling of nuclei

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The dynamics of nuclear spins in conditions of nuclear dynamic cooling is investigated. The process of establishment of the stationary polarization is analyzed for various intensities of the alternating magnetic field. An equation describing the nuclear polarization process in the case of a strong saturating field at low temperatures is obtained.

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In recent years the introduction of the concept of a spin–spin interaction reservoir has turned out to be extremely fruitful in the development of magnetic resonance in solids.[1–3] According to this concept the spin-spin interaction energy (more precisely, its secular part) is regarded as a separate energy reservoir, isolated, generally speaking, from the Zeeman energy of the spins in the external magnetic field $\mathbf{H}_0$ and characterized by its own temperature, which, under certain conditions, can differ greatly from the Zeeman temperature.

In Refs. 2 and 3, the existence of thermal contact between the dipole reservoir of paramagnetic impurities and the Zeeman system of the nuclei[4] was predicted theoretically, and was later confirmed in numerous experiments. In the presence of near-resonance saturation of the EPR the temperature of the dipole reservoir of the electron spins is lowered. The presence of the effective coupling with the Zeeman system of the nuclei leads to lowering of the nuclear Zeeman temperature too, and this increases the nuclear polarization. This method of polarization has been named the "method of dynamics cooling of nuclei." A number of theoretical and experimental papers[5] are devoted to the study of this method. The method of dynamic cooling of nuclei

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