Admissible values of permittivity and magnetic permeability of matter

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Results are presented of a systematic investigation of an equilibrium homogeneous and isotropic medium from the viewpoint of its causal properties (the Kramers-Kronig and Leontovich relations) and of its stability to electromagnetic perturbations. The ranges of permissible values of the permittivity and of the magnetic permeability of matter are then found in the static limit and over the entire range of variation of the wave vector. It is shown that the magnetic permeability, unlike the permittivity, cannot be negative; that the lower limit of the magnetic permeability (which coincides with the magnetic permeability of a London superconductor) increases with increasing wave vector and approaches unity, that no order parameter **H** (magnetic field) can be produced via a phase transition from the state of a stable homogeneous and isotropic medium, and that in this sense no media with spontaneous homogeneous electric field can exist.

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1. INTRODUCTION

Several general questions in electrodynamics of continuous media are still not clear enough, even though they are of interest not only from the viewpoint of general physics, but are also closely related with timely physics problems such as that of high-temperature superconductivity,[1] anomalous diamagnetism and spontaneous currents,[2] physical aspects of color containment in chromodynamics,[3] and others.

It is convenient to formulate these questions in the language of the static ($\omega = 0$) values of the permittivity $\varepsilon(\omega,\mathbf{k})$ and of the magnetic permeability $\mu(\omega,\mathbf{k})$ of an equilibrium homogeneous and isotropic medium.

a) In vacuum, where ε and μ are positive, like charges repel and currents flowing in the same direction attract each other. Are there any media in which the situation is reversed (ε or μ is negative)?

b) In the long-wave limit $\mathbf{k} \to 0$ there exist paraelectrics ($\varepsilon > 1$), magents ($\mu > 1$), and diamagnets ($0 < \mu < 1$), whereas no diaelectrics ($0 < \varepsilon < 1$) are possible. Does the situation change in this respect when the wave vector $\mathbf{k}$ is increased (does diaelectricity appear and does diamagnetism vanish in this case)?

c) An electric order parameter can be either an electric field $\mathbf{E}$ (charge-density waves, two-stream instability, etc., which occur at $\varepsilon = 0$) or the induction (ferroelectricity, $\varepsilon \to \infty$). Yet the only known order parameter in magnetically ordered media is the magnetic induction $\mathbf{B}$ (ferro- and antiferromagnets, $\mu \to \infty$). Can media exist in which the order parameter is the field $\mathbf{H}$ (diamagnetic spontaneous currents, $\mu = 0$)?

d) The order parameter of a ferroelectric or a ferromagnet is homogeneous in space $\mathbf{k} = 0$, whereas the known media with order parameter $\mathbf{E}$ [see subsec. (c)] have $\mathbf{k} \neq 0$. Can media whose order parameter is a homogeneous electric field exist?

These and more particular questions of the same kind reduce in the upshot to the following question:

e) What is the range of admissible values of ε and μ of an equilibrium medium in the entire range of $\mathbf{k}$, and what is the physical meaning of the boundaries of this region.

In the literature the answer to this question is as a rule incomplete and sometimes simply incorrect (see, however, the article by Martin[4] and its criticism below). In the last few years two of us, jointly with Maximov, determined in a number of papers (Ref. 5, see also Ref. 1) the range of permissible values of ε and discussed the ensuing consequences as applied to the problem of high-temperature superconductivity. The present article, a development of the cited ones, is an attempt to provide as full and as clear-cut answers as possible to the foregoing questions.

We consider below only homogeneous and isotropic media of sufficiently large size. Accordingly, the results that follow are not directly applicable to ordered media with an electromagnetic order parameter that singles out directions and points in space. The medium is assumed to be nonrelativistic to the extent compatible with the existence of magnetism itself. The restrictions above are assumed only for the sake of simplicity, and in later publications we shall consider a more general situation.

The plan of the exposition is the following. In Sec. 2 we introduce the material equations that will be useful later on. We discuss next the electromagnetic response functions (Sec. 3) and formulate dispersion relations (Sec. 4) from which we deduce restrictions on the permissible values of ε and μ (Sec. 5). In Sec. 7 the same restrictions are obtained from a material-stability condition formulated in Sec. 6. In the concluding Sec. 8 we discuss the results and answer the questions posed above.

2. MATERIAL EQUATIONS

There exist several methods of introducing the material equations that describe the properties of a medium and make the system of Maxwell's equations closed. In addition to the

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most widely used ones, we indicate below a third method, more convenient from the viewpoint of interest to us.

We consider hereafter only a monochromatic case and omit in most equations the arguments ω and $\mathbf{k}$. Maxwell's equations for vacuum relate the corresponding fields $\mathbf{E}^e$ and $\mathbf{H}^e$ with the densities ρ^e and $\mathbf{j}^e$ of the external charge and of the current:

$$[\mathbf{k}\times\mathbf{H}^e]+\omega\mathbf{E}^e=-4\pi i\mathbf{j}^e,\quad \mathbf{k}\mathbf{H}^e=0,$$
$$[\mathbf{k}\times\mathbf{E}^e]-\omega\mathbf{H}^e=0,\quad \mathbf{k}\mathbf{E}^e=-4\pi i\rho^e. \tag{2.1}$$

Maxwell's equation in a medium are

$$[\mathbf{k}\times\mathbf{B}]+\omega\mathbf{E}=-4\pi i\mathbf{j},\quad \mathbf{k}\mathbf{B}=0,$$
$$[\mathbf{k}\times\mathbf{E}]-\omega\mathbf{B}=0,\quad \mathbf{k}\mathbf{E}=-4\pi i\rho, \tag{2.2}$$

where $\rho=\rho^i+\rho^e$ and $\mathbf{j}=\mathbf{j}^i+\mathbf{j}^e$, while ρ^i and $\mathbf{j}^i$ are the densities of the induced charge and current produced by the action of ρ^e and $\mathbf{j}^e$.

It is customary to introduce the material equations by changing from ρ^i and $\mathbf{j}^i$ to the vectors $\mathbf{D}$ and $\mathbf{H}$ and by establishing the connections between the vectors $\delta\,\mathbf{D}$ and $\delta\,\mathbf{E}$ and between $\delta\,\mathbf{B}$ and $\delta\,\mathbf{H}$ (δ is the small-variation symbol). Equations (2.2) themselves then take the form

$$[\mathbf{k}\times\mathbf{H}]+\omega\mathbf{D}=-4\pi i\mathbf{j}^e,\quad \mathbf{k}\mathbf{B}=0,$$
$$[\mathbf{k}\times\mathbf{E}]-\omega\mathbf{B}=0,\quad \mathbf{k}\mathbf{D}=-4\pi i\rho^e. \tag{2.3}$$

It is important that $\mathbf{D}$ and $\mathbf{H}$ are not uniquely defined, with a leeway of two degrees of freedom (the six of their components are subject, with account taken of the continuity equation, to four conditions—Eqs. (2.3)). It is this leeway which accounts for the different methods of introducing the material equations (see Ref. 6 in this connection).

They can be formulated, for example, so that the vectors $\delta\,\mathbf{D}$ and $\delta\,\mathbf{H}$ are directed respectively along $\delta\,\mathbf{E}$ and $\delta\,\mathbf{B}$:

$$\delta\mathbf{E}=\varepsilon^{-1}\delta\mathbf{D},\quad \delta\mathbf{B}=\mu\delta\mathbf{H}, \tag{2.4}$$

where ε and μ are the permittivity and the magnetic permeability referred to in the Introduction. Another frequently used method consist of subjecting $\delta\,\mathbf{H}$ to the condition $\delta\,\mathbf{H}=\delta\,\mathbf{B}$. This yields

$$\delta\mathbf{E}_\parallel=\varepsilon_\parallel^{-1}\delta\mathbf{D}_\parallel,\quad \delta\mathbf{E}_\perp=\varepsilon_\perp^{-1}\delta\mathbf{D}_\perp. \tag{2.5}$$

The subscripts $\parallel$ and $\perp$ mark respectively the components longitudinal and transverse relative to $\mathbf{k}$. The magnetic properties of the medium are described by the quantity $\varepsilon_\perp$ connected in a known manner with ε and μ.[7,8]

Comparison of (2.3) with (2.1) shows that at a planar geometry of the problem $\mathbf{D}_\parallel$ coincides with the external field $\mathbf{E}^e_\parallel$. It would be very convenient (see below) if $\mathbf{H}$ and $\mathbf{H}^e$ were also to coincide. For the second method this patently not the case, and for the first case this is valid only at $\omega=0$, by virtue of the easily verified equality

$$\mathbf{H}=\mathbf{H}^e\frac{\omega^2-k^2}{\omega^2\varepsilon\mu-k^2}.$$

It is therefore desirable to use a third method of introducing the material equations, wherein $\delta\,\mathbf{H}=\delta\,\mathbf{H}^e$ at all frequencies. Corresponding to it are the equations

$$\delta\mathbf{E}_\parallel=\alpha\delta\mathbf{E}_\parallel{}^e,\quad \delta\mathbf{B}=\beta\delta\mathbf{H}^e, \tag{2.6}$$

or, equivalently,

$$\delta\rho=\alpha\delta\rho^e,\quad \delta\mathbf{j}_\perp=\beta\delta\mathbf{j}_\perp{}^e. \tag{2.6$'$}$$

The quantities α and β in these equations describe the medium just as completely as ε and μ or $\varepsilon_\parallel$ and $\varepsilon_\perp$. The connection between all of them is given by Eqs. (2.1)–(2.3):

$$\alpha=1/\varepsilon=1/\varepsilon_\parallel,\quad \beta=\frac{\omega^2-k^2}{\omega^2\varepsilon-k^2\mu^{-1}}=\frac{\omega^2-k^2}{\omega^2\varepsilon_\perp-k^2}. \tag{2.7}$$

It can be seen that at $\omega=0$ the value of β coincides with the static magnetic permeability, and as $\omega\to\infty$ it coincides with $1/\varepsilon_\perp$. An additional advantage of this method is that β, in contrast to the magnetic permeability, has a direct physical meaning at all frequencies and, unlike $\varepsilon_\perp$, has no pole at $\omega=0$.

3. RESPONSE FUNCTIONS

We subject to medium in question to a small external action (I) that does not necessarily stem from external charges or currents. The result of the action is a quantity A that characterizes the changes produced in the medium and is connected with I by the relation

$$A=R\times I, \tag{3.1}$$

where the response function R describes the reaction of the medium to the external action regardless of its amplitude. The material equations (2.4)–(2.6) considered have just the form of this relation.

For R to be indeed the response function (and satisfy the causality condition that will be important in what follows, see Sec. 4 below) it is necessary that the action I be truly external, i.e., that it experience no reaction from the medium itself and by the same token be controllable and capable of assuming any prescribed value.[9] This requirement, the necessity of which as applied to electrodynamics of continuous media was emphasized by Pines and Nozières,[10] is far from always satisfied; this indeed was the source of widespread incorrect conclusions in the literature. Our immediate task is therefore to ascertain which of the quantities $\varepsilon,\mu,\varepsilon_\parallel,\varepsilon_\perp,\alpha,\beta$ and their inverses can be regarded as response functions, and under which conditions.

We distinguish next between two values of the vector $\mathbf{k}$ in connection with Eq. (3.1). The first, $\mathbf{k}=0$ (more accurately $k\lesssim L^{-1}$, where L is the large linear dimension of the medium) can be realized by sources located outside the medium and whose field has as $L\to\infty$ only a homogeneous component. In the second case, action with $\mathbf{k}\neq0$, the sources must be placed inside the medium. This difference is important from the viewpoint of the reaction of the medium on the action source.

The condition to which I must be subjected is satisfied in any case by the fields $\delta\,\mathbf{E}^e$ and $\delta\,\mathbf{H}^e$ generated by prescribed external charges and currents. At $\mathbf{k}=0$ such an action is produced by charges on capacitor electrodes between which the medium is placed (longitudinal electric field, Fig. 1a), or else by a current in the winding of a solenoid that envelopes the medium (magnetic field, Fig. 1b). On the other

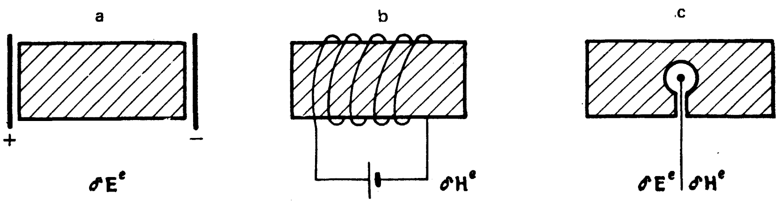

FIG. 1.

hand an action with $\mathbf{k}\neq 0$ can be produced by introducing into the system a probe with small controllable charge or current on its end (Fig. 1c). The corresponding densities have all the Fourier components with respect to $\mathbf{k}$.

From this and from a comparison of (2.6) with (3.1) it follows that the quantities α (and simultaneously also $1/\varepsilon$ and $1/\varepsilon_{\parallel}$) and β can be regarded as response functions at all values of $\mathbf{k}$:

$$\alpha=R, \tag{3.2a}$$

$$\beta=R. \tag{3.2b}$$

However, the customarily employed quantities μ and $1/\varepsilon_{\perp}$ are not response functions, since the corresponding quantities $\delta\mathbf{H}$ and $\delta\mathbf{D}_{\perp}$ are not controllable, being dependent on the state of the medium at $\omega\neq 0$ (see Sec. 2). Thus is precisely the advantage of the method introduced in Sec. 2 for formulating the material equations.

In the situation considered the role of I was played by the external fields $\delta\mathbf{E}^e$ and $\delta\mathbf{H}^e$, and that of A by the total fields $\delta\mathbf{E}$ and $\delta\mathbf{B}$. The inverse situation, when the external and total field change places, is also possible, but only at $\mathbf{k}=0$, when the sources I are located outside the system. For a longitudinal electric field this is a capacitor to whose electrodes are applied a controllable charge, as above, and a controllable potential difference (Fig. 2a). For a magnetic field this is a superconductor in the cavity of which is placed the medium (Fig. 2b). Since the flux of the field $\mathbf{B}$ through the cavity is quantized, $\mathbf{B}$ does not depend on the state of the medium and can be controlled by varying the area of the cavity. The role of A, on the other hand, is assumed in this case by the fields of the external charge that flows to the electrodes or flows away from them, or of an external current flowing over the inner surface of the superconductor. Accordingly, at $\mathbf{k}=0$ the quantities $1/\alpha$ (and simultaneously ε and $\varepsilon_{\parallel}$) and $1/\beta$ ($\mathbf{k}=0$) are also response functions:

$$1/\alpha=R, \tag{3.3a}$$

$$1/\beta=R. \tag{3.3b}$$

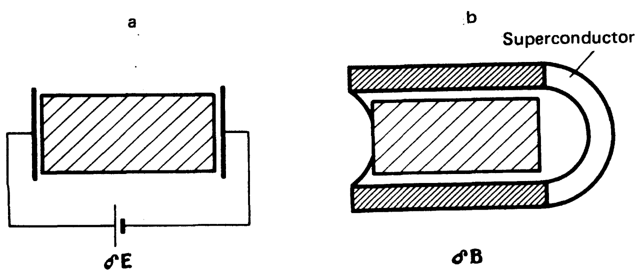

FIG. 2.

As for action with $\mathbf{k}\neq 0$ by the total fields, it cannot be realized. The point, in the upshot, is that the ease of controlling the specified external field is conteracted by the difficulty of controlling the total fields, which contain an uncontrollable contribution of the medium itself.[10] The distinct character of the case $\mathbf{k}=0$ for the response to a total field will be illustrated in Sec. 7 below from the viewpoint of the stability of the medium.

4. DISPERSION RELATIONS

The response function satisfies the causality condition. In the case of a longitudinal electric field we can confine ourselves, as in the papers cited above, to the nonrelativistic causality condition: the result of the action A is zero at instants of time preceding the action I itself. From this follows the known conclusion that the response function $R(\omega,\mathbf{k})$ is analytic as a function of the frequency in the upper ω half-plane at all values of $\mathbf{k}$. This makes it possible to apply the Cauchy formula to the function $R(\omega,\mathbf{k})-R(\Omega,\mathbf{k})$ (here and below $\Omega\to\infty$), and this leads to a relation of the Kramers-Kronig type (see, e.g., Ref. 10):

$$R(\omega,\mathbf{k})=R(\Omega,\mathbf{k})+\frac{1}{\pi}\int_0^\infty\frac{\operatorname{Im}R(\omega',\mathbf{k})}{\omega'^2-\omega^2-i\delta}\,d\omega'^2. \tag{4.1}$$

In the case, however, of a magnetic field that is relativistic in character, relation (4.1) yields too little information. It is necessary to turn to the more stringent relativistic causality condition, which requires that A be zero in those cases when the events A and I are connected by a space-like interval that cannot be overcome by a signal moving not faster than light. This leads to dispersion relations that are more restrictive than (4.1). They were pointed out by Leontovich[11] (see also Ref. 8). The relativistic causality condition leads to analyticity of the system of functions $R(\omega',\mathbf{k}')$, where ω' and $\mathbf{k}'$ are the frequency and the wave vector in a reference frame that moves relative to the initial one at a velocity $\mathbf{u}$ that takes on all values from zero (nonrelativistic causality condition) to unity (speed of light). From this we can arrive at the relation

$$R(\omega,\mathbf{k})=R(\Omega,\mathbf{k}-\Omega\mathbf{u})+\frac{1}{\pi}\int_0\frac{\operatorname{Im}R(\omega',\mathbf{k}-\mathbf{u}(\omega'-\omega))}{\omega'^2-\omega^2-i\delta}\,d\omega'^2. \tag{4.2}$$

It leads to the strongest restrictions if one chooses

$$\mathbf{uk}=0,\quad u=1, \tag{4.3}$$

and since R in a homogeneous isotropic medium depends only on the modulus of $\mathbf{k}$, the matter reduces to a substitution of the type $\mathbf{k}-\mathbf{u}\omega\to(k^2+\omega^2)^{1/2}$. Accordingly, the static ($\omega=0$) limits of relations (4.1) and (4.2), in which we shall be interested, take the form

$$R(0,\mathbf{k})=R(\Omega,\mathbf{k})+\frac{1}{\pi}\int_0^\infty \frac{d\omega'^2}{\omega'^2}\,\mathrm{Im}\,R(\omega',\mathbf{k}), \tag{4.4}$$

$$R(0,\mathbf{k})=R(\Omega,(k^2+\Omega^2)^{1/2})+\frac{1}{\pi}\int_0^\infty \frac{d\omega'^2}{\omega'^2}\,\mathrm{Im}\,R(\omega',(k^2+\omega'^2)^{1/2}). \tag{4.5}$$

From this we can find the permissible values of $R(0,\mathbf{k})$ if we know the terms outside the integrals and the signs of the integrands in the right-hand sides of (4.4) and (4.5).

Taking next R to mean the quantities α, β, and their reciprocals (see (3.2) and (3.3)), we obtain the signs of their imaginary parts. The known expression for the energy dissipated by a monochromatic wave per unit time[7,8]

$$Q=\frac{\omega}{8\pi}[\mathrm{Im}\,\varepsilon_\parallel|E_\parallel|^2+\mathrm{Im}\,\varepsilon_\perp|E_\perp|^2]\geqslant 0$$

yields the inequalities $\mathrm{Im}\,\varepsilon_\parallel\geqslant 0$ and $\mathrm{Im}\,\varepsilon_\perp\geqslant 0$. From them and (2.7) we get

$$\mathrm{Im}\,\alpha\leqslant 0, \tag{4.6a}$$

$$(\omega^2-k^2)\,\mathrm{Im}\,\beta\leqslant 0. \tag{4.6b}$$

The imaginary parts of the reciprocals are of opposite sign.

The asymptotes of α and β as $\omega\to\infty$ follow from the fact that the medium does not manage to react to an action whose frequency exceeds its characteristic frequencies (remaining, of course, lower than the rest mass of the particles of the medium). In this limit the corresponding increments to the particle wave function vanish and, as a consequence, the increments to the induced charge and to the paramagnetic current (see below). Therefore as $\omega\to\infty$ the response functions will be the same as for vacuum and, in particular,

$$\alpha(\Omega,\mathbf{k})=1. \tag{4.7}$$

Another asymptotic expression we need is

$$\beta(\Omega,(\Omega^2+k^2)^{1/2})=(1+\omega_p^2/k^2)^{-1}, \tag{4.8}$$

where ω_p^2 is the sum, over all the charged-particle species, of the squares of their plasma frequencies $4\pi e\rho/m$ (e is the charge, ρ is its density, and m is the particle mass). For a rigorous derivation of (4.8) see the Appendix, a simplified one is given below.

Starting with the second Eq. (2.6), we have $\delta\mathbf{j}=\delta\mathbf{j}^p+\delta\mathbf{j}^d+\delta\mathbf{j}^e$ (the index $\perp$ will be omitted from now on), where the paramagnetic $\mathbf{j}^p$ and the diamagnetic $\mathbf{j}^d$ current components are of the form

$$\mathbf{j}^p=\frac{e}{2m}[i(\nabla\bar\psi\psi-\bar\psi\nabla\psi)+\mathrm{rot}(\bar\psi\boldsymbol{\sigma}\psi)],$$

$$\mathbf{j}^d=-\frac{e\rho}{m}\mathbf{A},\qquad \rho=e\bar\psi\psi$$

(ψ is the particle wave function, $\boldsymbol{\sigma}$ is the spin operator, the summation over the particles was left out). It must be emphasized that the diamagnetic part of the current contains the *total* vector potential, which satisfies the equation $(\omega^2-k^2)\mathbf{A}=4\pi\mathbf{j}$ and includes the induced-current field. This simple fact, which is obvious, e.g., from the viewpoint of the general equations of quantum electrodynamics, is ignored in many books. Taking all the foregoing into account[1)] we have

$$\delta\mathbf{j}=\boldsymbol{\xi}+[1+\omega_p^2/(k^2-\omega^2)]^{-1}\delta\mathbf{j}^e,\quad \boldsymbol{\xi}=[1+\omega_p^2/(k^2-\omega^2)]^{-1}\delta\mathbf{j}^p \tag{4.9}$$

(ρ is not altered by transverse action). As $\omega\to\infty$ we have $\delta\mathbf{j}^p\to 0$ (see above) and from (4.9) and (2.6) we get (4.8).

5. ADMISSIBLE VALUES OF THE PERMITTIVITY AND OF THE MAGNETIC PERMEABILITY

The results enable us to obtain the admissible static values of the permittivity and of the magnetic permeability. For a longitudinal electric field, Eqs. (3.2a), (4.6a), (4.7), and (4.4) lead to the inequality

$$\alpha(0,\mathbf{k})=1/\varepsilon(0,\mathbf{k})\leqslant 1\quad(\mathbf{k}-\text{arbitrary}), \tag{5.1}$$

and the use of (3.3a) in place of (3.2a) yields

$$1/\alpha(0,\mathbf{k})=\varepsilon(0,\mathbf{k})\geqslant 1\quad(\mathbf{k}=0). \tag{5.2}$$

A detailed discussion of the consequences of these equations (in particular, of the question of media with $\varepsilon(0,\mathbf{k})<0$) is contained in Ref. 5.

For a magnetic field, the Leontovich dispersion relation leads to stronger restrictions than the Kramers-Kronig relation. At the same time, it is more convenient, since the first factor in (4.6b) is of definite sign if the vector $\mathbf{u}$ is chosen in the form (4.3). For the response to an external field [relations (3.2b), (4.6b), (4.8), (4.5)] we have

$$\beta(0,\mathbf{k})=\mu(0,\mathbf{k})\geqslant(1+\omega_p^2/k^2)^{-1}\quad(\mathbf{k}-\text{arbitrary}), \tag{5.3}$$

and for the response to the total field [(3.3b) in place of (3.2b)] we have

$$1/\beta(0,\mathbf{k})=1/\mu(0,\mathbf{k})\leqslant 1+\omega_p^2/k^2\quad(\mathbf{k}=0). \tag{5.4}$$

The Kramers-Kronig relation, however, yields for the response to the total field the same result (5.4), while for the rsponse to an external field it yields an inequality weaker than (5.3), viz.,[2)]

$$\mu(0,\mathbf{k})\geqslant 1-\omega_p^2/k^2. \tag{5.5}$$

As $c\to\infty$, when the formulations of the causality conditions that are the basis of (5.3) and (5.5) coincide and the inequalities themselves also coincide (in standard units the factor ω_p^2/k^2 in these inequalities takes the form ω_p^2/k^2c^2). We note in this connection that retention of terms of higher order in $1/c^2$ in (5.3) is not an exaggeration of the accuracy within the framework of the nonrelativistic calculation: these terms correspond to the parameter ω_p^2/k^2c^2, which is not small (at small $\mathbf{k}$), whereas higher powers of the small parameter v^2/c^2, where $\mathbf{v}$ is the velocity of the particles of the medium, were discarded in the expression for the current (see above).

The inequalities obtained are illustrated in Fig. 3, where the regions of the admissible values of the permittivity (Fig. 3a) and of the magnetic permeability (Fig. 3b) are shaded, and the dashed lines denote the restrictions (5.5). It can be seen that for the permittivity the response to the total field leads to stronger restrictions that pertain, however, to the point $\mathbf{k}=0$. For the magnetic permeability, on the contrary, stronger restrictions are imposed by the response to an external field, which pertains furthermore to the entire range of $\mathbf{k}$. A more detailed discussion of the consequences of Eqs. (5.3) and (5.4) is contained in the Conclusion.

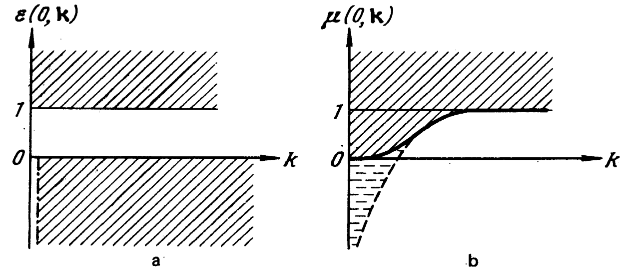

FIG. 3.

We note that the inequalities discussed can be arrived at on the basis not of the dispersion relations but of the essentially equivalent Kubo relations that express the generalized susceptibility in terms of the retarded commutator of the corresponding currents.[12] This procedure leads, in particular, to a known relation that yields (5.1) directly:

$$1/\varepsilon(0,\mathbf{k})=1-\frac{8\pi}{k^2}\sum_n |\langle n|\rho^i(\mathbf{k})|0\rangle|^2/\omega_{n0}, \qquad (5.6)$$

where $\omega_{n0}=E_n-E_0$, with E_n and E_0 the excited and ground energy levels, respectively. A similar formula for μ, leading to (5.3), can be obtained from the results of the Appendix (this formula differs from (5.6) in that unity is replaced by $(1+\omega_p^2/k^2)^{-1}$ and ρ^i is replaced by ξ, see (4.9)).

This method was used by Martin in his cited paper,[4] where inequalities similar to (5.1)–(5.4) are given. Their derivation, however, is highly unsatisfactory not only because they are formal and unaccompanied by a physical discussion (no notice is taken, for example, of the feasibility of negative permittivity), but also because of shortcomings in substance. There is, for example, no argument at all in favor of the transition from inequality (5.4), which admits the possibility of $\mu<0$, to the stronger inequality, which obscures this possibility; the fact that the quantity $\sigma^{T\,\mathrm{reg}}$ is of definite sign, an important factor in the derivation, is not discussed; and so on. At the same time we must not overlook the great importance of this paper, where it was first stated that the Kramers-Kronig relation may be violated for the permittivity (case of response to the total field).

6. STABILITY OF MEDIUM

The question of satisfaction of the dispersion relation is closely connected with the question of the stability of the medium. We shall verify below directly that the inequalities (5.1)–(5.4) obtained above with the aid of the dispersion relations consititute none other than criteria of the stability of the medium to spontaneous onset and growth of electromagnetic-field fluctuations. Besides additionally confirming the results cited above, it becomes possible to understand, from a different viewpoint, why the inequalities (5.1) and (5.3) pertain to all values of $\mathbf{k}$ while (5.2) and (5.4) pertain only to the point $\mathbf{k}=0$.

The connection between the dispersion relations (causality condition) and the stability is based on simple physical considerations. In a medium that is unstable to the appearance and growth of a physical quantity A at a fixed—in particular, zero—value of the quantity I, the response function must have a pole (or a cut) in the upper frequency half-plane, meaning violation of the dispersion relations. Only in this case does the quantity A have an exponential growth with a rate determined by the imaginary part of the frequency at the pole. On the other hand, violation of the dispersion relations (with satisfaction, of course, of the physical causality principle) is possible only in the case of an unstable medium, where a nonzero value of A is produced spontaneously, unconnected at all with the action I. If this takes place prior to the start of the action I, we encounter an imitation of causality violation and nonsatisfaction of the dispersion relations.[9]

Proceeding now to formulation of conditions for stability of the medium with respect to the appearance and growth of fluctuations of a physical quantity Φ, we introduce the energywise conjugate external action—the "current" J. This means that variation of the free energy $F(J)$ (or simply of the energy at the temperature $T=0$) of the medium in an external field is equal to $\delta F(J)=\Phi\delta J$. We seek the free energy $F(\Phi)$ as a function of Φ (this is just the quantity on which the Landau phase-transition theory[12] is based), whose minimum determines the stable state of the system. The corresponding minimum conditions are in fact the conditions for the stability of the medium.

It is well known that a transition from the variable J to Φ is effected by a Legendre transformation

$$F(\Phi)=F(J)-J\Phi,\quad \delta F(J)/\delta J=\Phi \qquad (6.1)$$

followed by replacement of J with Φ with the aid of the second of these equations. Hence $\delta F(\Phi)=-J\delta\Phi$ and the stable state of the medium is determined by the conditions

$$\frac{\delta F(\Phi)}{\delta\Phi}=-J(\Phi)=0,\quad \frac{\delta^2F(\Phi)}{\delta\Phi^2}=-\frac{\delta J(\Phi)}{\delta\Phi}=-\left(\frac{\delta\Phi(J)}{\delta J}\right)^{-1}\geqslant 0. \qquad (6.2)$$

The first of them corresponds to the absence of external action (the medium is represented by itself), and the second is the stability criterion proper. We note that physically the inequality (6.2) corresponds to the known Le-Chatelier–Braun principle [12,13]: the changes produced in a stable medium by an external action are such that they decrease the consequences of this action (see also Ref. 5).

The Legendre transformations (6.1) correspond to the physical picture frequently called the Leontovich principle.[14] It is based on the following considerations. We are dealing with an expression for the function $F(\Phi)$ at arbitrary, including nonequilibrium, values of Φ in terms of the purely equilibrium function $F(J)$. This is possible if the current J is chosen such that the given value of Φ becomes equilibrium. In this case, however, we deal not with a free medium left to itself, but with a medium situated in an external field J. Therefore we must subtract from the free energy $F(J)$ the "extra" work corresponding to the energy in this field [the subtracted term in the Legendre transformation (6.1)].

It is important to emphasize that in the presence of an external field the quantity Φ itself will also contain an "extra" component due to this field. This component, which we designate Φ_j, must also be subtracted from the corresponding expression. Therefore, to obtain the correct stability criterion, it is necessary either to define Φ from the very outset in such a way that it contain no extra terms, or replace the second condition of (6.2) by the inequality

$$\delta(\Phi(J)-\Phi_J)/\delta J\leqslant 0. \tag{6.3}$$

Only then are we actually investigating the stability of a free medium, not subject to the action J, against spontaneous appearance and growth of the corresponding fluctuations. All the foregoing will be illustrated in the next section, using the electromagnetic field as an example.

7. CRITERIA OF ELECTROMAGNETIC STABILITY

The criterion of the stability of a medium to the growth of fluctuations of an electromagnetic field will be expressed below in terms of the response functions α and β. Since these criteria pertain to an equilibrium state of the medium, the answer is expressed in terms of the static values $\alpha(0,\mathbf{k}) = 1/\varepsilon(0,\mathbf{k})$ and $\beta(0,\mathbf{k}) = \mu(0,\mathbf{k})$, and the derivation itself can be carried out within the framework of electrostatics and magnetostatics.

We consider first stability with respect to the appearance of an electric field $\mathbf{E}$ (the quantity A) in the absence of induction $\mathbf{D}$ (or, equivalently, of the field $\mathbf{E}^e$), which assumes the role of I; the response function is the quantity $\alpha(0,\mathbf{k})$. This corresponds physically to Figs. 1a and 1c when the plates and the probe are uncharged (or even absent). Starting from the known relation[15] $\delta F = \mathbf{E}\delta\mathbf{D}/4\pi$, we can identify the current J with the quantity $\mathbf{D}/4\pi$, and Φ with $\mathbf{E}$. In accord with the statements made at the end of Sec. 6, however, the quantity $\mathbf{E} = \mathbf{D} - 4\pi\mathbf{P}$ ($\mathbf{P}$ is the polarization of the medium) contains the term $\Phi_J = \mathbf{D}$. Therefore, according to (6.3), the stability criterion takes the form

$$\delta(\mathbf{E}-\mathbf{D})/\delta\mathbf{D}=\alpha(0,\mathbf{k})-1\leqslant 0, \tag{7.1}$$

which coincides with (5.1).

The investigation of the stability of the medium with respect to the appearance of the induction $\mathbf{D}$ (the result A) at a zero field $\mathbf{E}$ (the action I) with a response function $1/\alpha(0,\mathbf{k})$ corresponds to Fig. 2a with grounded or short-circuited capacitor plates. The relation $\delta F = -\mathbf{D}\delta E/4\pi$ (Ref. 15) makes it possible to set the current J in correspondence with $\mathbf{E}/4\pi$, and Φ with $-\mathbf{D}$; the latter, represented in the form $-\mathbf{D} = -\mathbf{E} - 4\pi\mathbf{P}$, contains the term $\Phi_J = -\mathbf{E}$ due to the external field. The inequality (6.3) yields the stability criterion

$$\delta(\mathbf{E}-\mathbf{D})/\delta\mathbf{E}=1-1/\alpha(0,\mathbf{k})\leqslant 0, \tag{7.2}$$

which coincides with inequality (5.2). Just as the latter, the condition (7.2) pertains only to the point $\mathbf{k} = 0$: the spontaneously produced induction satisfies inside the medium the equation $\mathbf{k}\cdot\mathbf{D} = 0$ [see (2.3)]. Actually, as we can see, in both cases we are dealing with spontaneous onset of polarization that plays in fact the role of the quantity $\Phi - \delta\Phi_J$ in (6.3).

It might seem that a similar role as applied to magnetic stability should be played by the magnetization $\mathbf{M} = (\mathbf{B} - \mathbf{H})/4\pi$. However, the criteria corresponding to (7.1) and (7.2) are such that they exclude the existence of diamagnetism. The point is that the magnetization itself contains a term proportional to the current J, which must also be subtracted from the expression for Φ. This term, diamagnetic in nature, is designated $\delta\Phi_J$, so that Eq. (6.3) now contains the quantity $\Phi - \Phi_J - \delta\Phi_J$.

Examination of the stability with respect to the appearance of induction (the result A) at $\mathbf{H} = \mathbf{H}^e = 0$ (the action I) with a response function $\beta(0,\mathbf{k})$ corresponds to Fig. 1b in the absence of current in the solenoid, or in the absence of the solenoid itself. The relation[15] $\delta F = -\mathbf{B}\delta\mathbf{H}/4\pi$ makes it possible to identify J with $\mathbf{H}/4\pi$, Φ with $-\mathbf{B} = -\mathbf{H} - 4\pi\mathbf{M}$, and Φ_J with $-\mathbf{H}$. However, using (4.9) and the equations

$$i[k\mathbf{M}]=\mathbf{j}^i,\quad i[\mathbf{k}\times\mathbf{B}]=4\pi\mathbf{j},\quad i[\mathbf{k}\times\mathbf{H}]=4\pi\mathbf{j}^e,$$

we can write two equivalent relations:

$$-4\pi\mathbf{M}=\omega_p^2(k^2+\omega_p^2)^{-1}\mathbf{H}-(1+\omega_p^2/k^2)^{-1}\mathbf{B}^p, \tag{7.3a}$$

$$-4\pi\mathbf{M}=(\omega_p^2/k^2)\mathbf{B}-\mathbf{B}^p, \tag{7.3b}$$

where we have introduced the paramagnetic induction $\mathbf{B}^p$, defined by the equation $i\mathbf{k}\times\mathbf{B}^p = 4\pi\mathbf{j}^p$. It can be seen from (7.3a) that the first term in the right-hand side is directly connected with the current J and must therefore be identified with $\delta\Phi_J$. This yields a criterion that coincides with (5.3)

$$\delta[\mathbf{H}/(1+\omega_p^2/k^2)-\mathbf{B}]/\delta\mathbf{H}=(1+\omega_p^2/k^2)^{-1}-\beta(0,\mathbf{k})\leqslant 0. \tag{7.4}$$

Finally, an investigation of the stability with respect to the appearance of a field $\mathbf{H}$ (response A) at $\mathbf{B} = 0$ (action I) with a response function $1/\beta(0,\mathbf{k})$ corresponds to Fig. 2b at a zero number of flux quanta through the opening in the superconductor. The relation $\delta F = \mathbf{H}\delta\mathbf{B}/4\pi$ yields

$$J=\mathbf{B}/4\pi,\quad \Phi=\mathbf{H}=\mathbf{B}-4\pi\mathbf{M},\quad \Phi_J=\mathbf{B},\quad \delta\Phi_J=(\omega_p^2/k^2)\mathbf{B}$$

(the right-hand side of (7.3b), which is proportional to J). This yields a criterion that coincides with (5.4):

$$\delta[\mathbf{H}-(1+\omega_p^2/k^2)\mathbf{B}]/\delta\mathbf{B}=1/\beta(0,\mathbf{k})-(1+\omega_p^2/k^2)\leqslant 0. \tag{7.5}$$

By virtue of the equation $\mathbf{k}\times\mathbf{H} = 0$, which is valid in the static case in the absence of a current $\mathbf{j}^e$ inside the system (see (2.3)), this criterion pertains only to the point $\mathbf{k} = 0$.

The physical nature of $\delta\Phi_J$ is simplest to explain using as an example the last of the considered cases. The equilibrium of the given value of $\mathbf{H}$ is ensured by inclusion of the current J, whose role is assumed by the quantity $\mathbf{B}$. However, by virtue of the equation $m\dot{\mathbf{v}} = e\mathbf{E} = -e\dot{\mathbf{A}}$, the very inclusion of the field $\mathbf{B}$ leads to the appearance of an "extra" (diamagnetic) current $-(e\rho/m)\mathbf{A}$, which in fact corresponds to the quantity $\delta\Phi_J$. The crucial point here is the induction law—turning on the magnetic field generates an electric field, which leads in fact to an additional "unwinding" of the charges. Nothing similar happens for a longitudinal field, and the corresponding quantity $\delta\Phi_J$, and with it diaelectricity, is absent. The ensuing difference between electricity and magnetism is more fundamental than the difference corresponding to linearity of the Hamiltonian relative to the scalar potential and its quadratic dependence on the vector potential,[15] all the mores since in the relativistic theory (the Dirac equation) the Hamiltonian is linear with respect to all the components of the potential.[3)]

To complete this section, we present explicit expressions for that part of the free energy $F(\Phi)$ which has at equilibrium a minimum relative to spontaneous appearance of the fields **E**, **D**, **B**, and **H**, and is the first term of the corresponding Landau-theory expansion[12]:

$$F(\mathbf{E})=\varepsilon(\varepsilon-1)\mathbf{E}^2/8\pi, \quad F_0(\mathbf{D})=(\varepsilon-1)\mathbf{D}^2/8\pi\varepsilon^2,$$

$$F(\mathbf{B})=[\mu-(1+\omega_p^2/k^2)^{-1}]\mathbf{B}^2/8\pi\mu^2,$$

$$F_0(\mathbf{H})=\mu[(1+\omega_p^2/k^2)\mu-1]\mathbf{H}^2/8\pi. \quad (7.6)$$

Here F is the free energy for arbitrary **k**, and F_0 is the free energy only for $\mathbf{k}=0$. We emphasize that these expressions must in no way be confused with the known expressions for the total free energy of a dielectric or a magnet in an external field (such as $\varepsilon\mathbf{E}^2/8\pi$ etc.), which correspond to quantities of the type $F(J)$ and which should not have a minimum in the equilibrium state. This confusion is the cause of incorrect statements that crept into many books concerning the permissible values of ε and μ.

8. CONCLUSION

Summarizing the analysis in this article, we point now to the answer to the questions posed in the Introduction, starting with the most general question (e).

e) The admissible range of variation of the permittivity and of the magnetic permeability is determined by Eqs. (5.1)–(5.4) and by Fig. 3 (it is more convenient to go over to Fig. 4, which shows the reciprocal quantities). The physical meaning of the boundaries of this region is the following. For the quantity $1/\varepsilon$ (Fig. 4a) the lower limit $\varepsilon=0$ at $\mathbf{k}\neq 0$ is the limit of stability with respect to spontaneous appearance of an electric field; the equality $\varepsilon(0,\mathbf{k}_0)=0$ means the appearance of a charge-density wave with a wave vector $\mathbf{k}_0$. The lower limit $\varepsilon\to\infty$ at $\mathbf{k}=0$ means the limit of the stability with respect to the appearance of homogeneous induction **D** (ferroelectricity.[4]) The upper limit $\varepsilon=1$ is not a stability boundary, but corresponds simply to the limiting state of an infinitely rarefied medium (vacuum).

For the quantity $1/\mu$ (Fig. 4b), the lower limit $\mu\to\infty$ is the stability boundary with respect to spontaneous appearance of magnetic induction **B** (ferromagnetism at $\mathbf{k}=0$ and antiferromagnetism at $\mathbf{k}\neq 0$). The upper limit $\mu=(1+\omega_p^2/k^2)^{-1}$, however, is not a stability boundary but corresponds to the limiting state of a London superconductor at absolute zero temperature (this state is not ordered in the electromagnetic sense—the Meissner current appears not spontaneously but is induced by an external field).

a) Media with negative ε (at $\mathbf{k}\neq 0$) actually exist (nonideal plasma, strong electrolytes, and others[5]). There are no media with negative μ.

b) With increasing **k**, no conditions appear for the onset of diaelectricity (at least in the nonrelativistic region), and the conditions for the onset of diamagnetism become more stringent (the lower limit of μ increases and approaches unity).

c) There are no states with a spontaneous magnetic field **H** (with spontaneous diamagnetic currents). They might be realized in the setup of Fig. 2b (just as ferroelectrics are realized in the setup of Fig. 2a) if an external current appears on the inner surface of the superconducting solenoid and is cancelled out, under the condition $\mathbf{B}=0$, by a spontaneous current in the medium. By virtue of $\delta\mathbf{B}=\mu\delta\mathbf{H}$, however, this calls for $\mu=0$ (ideal diamagnetism), and this is impossible at $\mathbf{k}\neq 0$ because of the restrictions obtained above on μ, and at $\mathbf{k}=0$ because of the Bloch theorem[16] (see also Ref. 2). We note incidentally that these restrictions do not exclude at all the existence of media with anomalously large diamagnetic susceptibility (see Fig. 3b). The very existence of such media, however, may be due, for example to a phase transition into a ferro- or antiferromagnetic state ($\mu\to\infty$).[2]

d) There are no media with spontaneous homogeneous electric field. They might occur by virtue of the equality $\delta\mathbf{D}=\varepsilon\delta\mathbf{E}$ at $\varepsilon=0$ at the pont $\mathbf{k}=0$, but this is prevented by the restrictions obtained above.

It should be noted, however, that the statements contained in (c) and (d) pertain, strictly speaking, only to the impossibility of the onset of a corresponding order parameter in a phase transition from the state of a stable homogeneous and isotropic medium on account of the appearance of instability to infinitely small perturbation. The problem itself of the existence of media with order parameters **H** and $\mathbf{E}=\text{const}$ is a broader one, and we hope to discuss it in the future.

We are grateful to V. L. Ginzburg and the participants of the seminar under his direction, particularly to E. G. Maksimov, for very helpful discussions.

APPENDIX

We present here a more rigorous derivation, than in the text, of the asymptotic (as $\omega\to\infty$ and at **k** either fixed or replaced by $\mathbf{k}-\mathbf{u}\omega$) equations

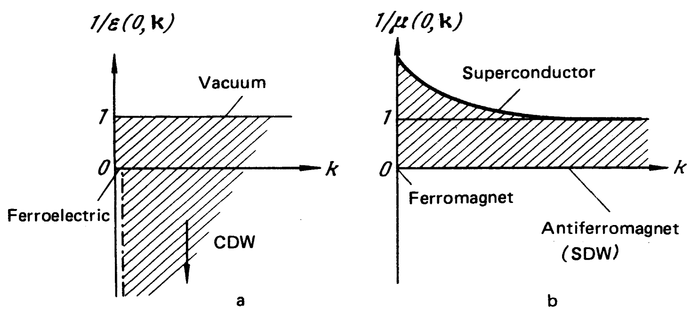

FIG. 4.

$$\delta\rho = \langle\delta\hat{\rho}\rangle \to \delta\rho^e \tag{A.1a}$$

$$\delta \mathbf{j} = \langle \delta\hat{\mathbf{j}}\rangle \longrightarrow \left[1+\frac{\omega_p^2}{k^2-\omega^2}\right]^{-1} \delta\mathbf{j}^e \tag{A.1b}$$

(the angle brackets denote averaging over the physical state). The operators of the charge density $\hat{\rho}$ and of the current $\hat{\mathbf{j}} = \hat{\mathbf{j}}^p + \hat{\mathbf{j}}^d + \mathbf{j}^e$ are expressed in terms of the Heisenberg operators $\hat{\psi}(\mathbf{x}, t)$ of the particle field and $\hat{\mathbf{A}}(\mathbf{x}, t)$ of the electromagnetic field in the form

$$\rho = e\hat{\psi}^+\hat{\psi},\ \hat{\mathbf{j}}^p = \frac{e}{2m}[i(\nabla\hat{\psi}^+\hat{\psi} - \hat{\psi}^+\nabla\hat{\psi}) + \mathrm{rot}(\hat{\psi}^+\sigma\hat{\psi})],$$

$$\hat{\mathbf{j}}^d = -\frac{e\hat{\rho}}{m}\hat{\mathbf{A}}.$$

From the equations of motion we obtain

$$\left[i\frac{\partial}{\partial t} - \frac{1}{2m}(\hat{\mathbf{p}} - e\hat{\mathbf{A}})^2 - \hat{U}\right]\hat{\psi} = 0, \quad \Box\hat{\mathbf{A}} = 4\pi\hat{\mathbf{j}} \tag{A.2}$$

($\hat{U}$ is the interaction expressed in terms of $\hat{\psi}^+$ and $\hat{\psi}$). The external action $\delta\mathbf{j}^e \sim \exp[i(\mathbf{kx} - \omega t)]$ adds to $\hat{\psi}$ and $\hat{\mathbf{A}}$ increments with an additional dependence on $\mathbf{x}$ and t in the form of the same exponential. Therefore when the first equation of (A.2) is solved by perturbation theory in $\delta\mathbf{j}^e$ we get the following replacements

$$i\partial/\partial t \to i\partial/\partial t + \omega, \quad \hat{\mathbf{p}} \to \hat{\mathbf{p}} + \mathbf{k}$$

and the addition

$$\delta\hat{\psi} \sim [\omega - k^2/2m + \ldots]^{-1} \quad \text{as } \omega \to \infty.$$

Accordingly as $\omega \to \infty$ the increment $\delta\hat{\rho}^i$ vanishes (this leads directly to (A.1a)), as does $\delta\hat{\mathbf{j}}_p$, so that we get $\delta\hat{\mathbf{j}} \to \delta\mathbf{j}^e - (e\hat{\rho}/m)\delta\bar{\mathbf{A}}$. Taking the second equation of (A.2) into account and making the substitutions $\hat{\rho} \to \rho + \hat{\rho}'$ and $\rho = \langle\hat{\rho}\rangle = \text{const}$, we get

$$\delta\mathbf{j} = \left(1 + \frac{\omega_p^2}{k^2-\omega^2}\right)^{-1} \delta\mathbf{j}^e + \delta\mathbf{j}',$$

where

$$\delta\mathbf{j}' = \left\{\left\langle\left[\frac{1+\left(\omega_p^2 + \dfrac{4\pi e}{m}\rho'\right)}{\left[(\mathbf{k}+\hat{\mathbf{p}})^2 - \left(\omega + i\dfrac{\partial}{\partial t}\right)^2\right]}\right]^{-1}\right\rangle - \left(1 + \frac{\omega_p^2}{k^2-\omega^2}\right)^{-1}\right\}\delta\mathbf{j}^e,$$

and the differentiation operators act on the fluctuating term $\hat{\rho}'$. It is easy to verify that $\delta\mathbf{j}'$ vanishes as $\omega \to \infty$ no more slowly than ω^{-2}, and this leads to (A.1b).

[1)]We note that relation (4.9) yields the correct spectrum of the transverse excitations of the medium $\omega^2 = \omega_p^2 + k^2$.

[2)]Its derivation is contained in V. V. Losyakov's diploma thesis, Moscow Physicotech. Inst., 1981.

[3)]Diamagnetism itself appears in the relativistic theory on account of virtual transitions into states with negative energy.

[4)]Antiferroelectrics, which correspond to growth of transverse lattice waves with $\mathbf{k} \neq 0$, can be shown not to be described in principle by the model of a homogeneous isotropic medium.

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