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Sum Rules, Kramers-Kronig Relations, and Transport Coefficients in Charged Systems

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A number of difficulties in previous treatments of the properties of linear homogeneous charged systems are discussed. With respect to the Kramers–Kronig relations and sum rules, it is argued that since the nonlocal electrical conductivity is the ratio of the current to the total field, a quantity which is physical but not necessarily arbitrary, the usual proof of the Kramers–Kronig relation fails. A valid proof, based on the dynamical properties of the electromagnetic field, is presented for the transverse electrical conductivity. On the other hand, it is shown by counterexample that there can be no general proof for the conventionally defined longitudinal electrical conductivity. Part of the difficulty may be ascribed to the failure of the conventional definition to account properly for the change in chemical potential which accompanies a longitudinal spatially varying field and charge distribution. A nonlocal quantity more closely related to the ratio of the longitudinal current to the longitudinal field plus the chemical potential gradient is shown to have more desirable causality properties. Rigorous expressions for the other transport coefficients (the thermopower and thermal conductivity) of simple charged systems are also obtained. These expressions differ substantially from the usually rigorous integral expressions associated with Kubo because of the long-range electromagnetic forces. A compilation of sum rules is included and a number of recurring pitfalls noted.

1. INTRODUCTION

DESPITE the large number of papers which have been written on the linear response of uniform homogeneous charged systems, surprisingly many facets of this theory have not been adequately or simply described. The purpose of this paper is to summarize this theory, distinguishing those aspects which are generally true from those aspects which are sometimes (or usually) true for some (or most) systems because of additional approximations which might well be violated in some physical systems. By generally true we mean that they follow for any homogeneous system of charged particles with magnetic moments, whose dynamics can be treated nonrelativistically, interacting with one another by nonelectromagnetic forces and by all the electromagnetic forces which Maxwell's equations imply. The homogeneity assumption (which we take to include translational and rotational invariance) prevents us from literally applying our conclusions to the ferromagnetic state, or to real metals, or even to models of superconductivity with no lattice when there is an equilibrium current J_s. Although most arguments can be generalized to include these effects, we shall not examine these generalizations here. We shall, however, take into account the magnetic interactions between particles, which are not, despite assertions to the contrary, of order $(v/c)^2$, since ferromagnetism hardly requires relativity. We also make no assumptions about the number of species or their mass ratio. Our results are therefore rigorous for plasmas and liquids (including liquid metals). Among the specific problems we discuss are the conditions under which the Kramers–Kronig relations apply, the conditions under which the sum rules for the electrical conductivity hold, the reconciliation of the Kramers–Kronig relations and sum rules with the properties of diamagnetic systems in general and superconductors in particular, and the rigorous expressions for transport coefficients in systems in which the long-range nature of the electromagnetic forces invalidates the usual Kubo correlation function definitions. Many of the subtleties we shall discuss are unlikely to concern the experimentalist. However, this paper contains some problems which may interest him. We point out, for example, that if he performs inelastic electron scattering experiments which determine $\mathrm{Im}[1/\epsilon^L(k\omega)]$ where $\epsilon^L(k\omega)$ is the conventionally defined wave number and frequency-dependent longitudinal dielectric constant, and from these, determines $\epsilon^L(k\omega)$, he may some day find that the quantities he has measured do not satisfy the familiar relations[1]

$$\mathrm{Re}[\epsilon^L(k\omega)-1]=\frac{2}{\pi}P\int_0^\infty d\omega'\,\frac{\mathrm{Im}\epsilon^L(k\omega')\omega'}{\omega'^2-\omega^2},$$

$$\mathrm{Im}\epsilon^L(k\omega)=-\frac{2}{\pi}P\int_0^\infty d\omega'\,\frac{\mathrm{Re}[\epsilon^L(k\omega')-1]\omega'}{\omega'^2-\omega^2},$$

or

$$\frac{2}{\pi}\int_0^\infty d\omega\,\mathrm{Im}\epsilon^L(k\omega)\omega=\frac{2}{\pi}\int_0^\infty d\omega\,\mathrm{Re}\sigma^L(k\omega)=\omega_p^2.$$

If he does, it would be interesting but he should not herald a breakdown of causality. A philosophically and physically respectable system may violate these equations, although most systems usually do not.

The author has also found that experimentalists, as well as some theorists, are unhappy about a much simpler question. How is it possible to reconcile orbital

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[1] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon Press, Inc., New York, 1960), Sec. 62.

diamagnetism, i.e., a negative static spin susceptibility $\chi_{MM}(0)$, with the Kramers–Kronig relation

$$\mathrm{Re}\chi_{MM}(\omega)=\frac{2}{\pi}P\int_0^\infty d\omega'\omega'\frac{\mathrm{Im}\chi_{MM}(\omega')}{\omega'^2-\omega^2}$$

and more explicitly

$$\mathrm{Re}\chi_{MM}(0)=\frac{2}{\pi}\int_0^\infty d\omega\frac{\mathrm{Im}\chi_{MM}(\omega)}{\omega}$$

and the requirement that $\mathrm{Im}\chi_{MM}(\omega)$ must be positive since it represents dissipation?[2] The experimentalist might also be interested in whether it is correct to assume that in homogeneous systems (a type-two superconductor with pinned flux lines is not homogeneous) superconductivity and the Meissner effect are equivalent. The answer is yes.

The casual reader will also no doubt be happy to hear that the local conductivity (i.e., $\sigma(0\omega)=i\omega[\epsilon(0\omega)-1]$) always satisfies the usual Kramers–Kronig relation and sum rule, and that the transverse nonlocal conductivity sum rule (which has been employed in superconductors[3]) is valid for all wave numbers even when magnetic effects are important (e.g., in a ferromagnetic material when the temperature is even infinitesimally above the Curie temperature). He will also not be upset to hear that there are rigorous expressions in terms of equilibrium correlation functions for the thermal conductivity and thermopower, even though the usual Kubo formulas[4] for these properties are incorrect because of the long-range nature of the Coulomb forces. Since it is possible that he may not be interested in the derivation of these expressions and properties, an attempt has been made to make the summary of sum rules (Sec. 7) comprehensible in terms of the standard definitions (3.1)–(3.5) and (4.1)–(4.6) to someone who has not read the intervening proofs in Secs. 3–6. The formulas for the dissipative coefficients are summarized at the end of Sec. 6.

2. CONVENTIONAL DISCUSSION OF KRAMERS-KRONIG RELATIONS AND SUM RULES FOR THE ELECTRICAL CONDUCTIVITY

In the discussion of electrical conductivity in plasmas it is conventionally stated that the measured, real part of the frequency-dependent conductivity $\sigma'(\omega)$ satisfies the sum rule

$$\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\sigma'(\omega)=\omega_p^2, \tag{2.1}$$

where ω_p is the plasma frequency. The sum rule is also stated for both the transverse and longitudinal conductivity even when the conductivity is nonlocal, that is, when the field varies sufficiently rapidly in space so that it is insufficient to assume that the conductivity

$$\sigma_{ij}(\mathbf{r}\mathbf{r}';\omega)=\sigma_{ij}'(\mathbf{r}\mathbf{r}';\omega)+i\sigma_{ij}''(\mathbf{r}\mathbf{r}';\omega), \tag{2.2}$$

which occurs in the linear relation

$$J_i(\mathbf{r}\omega)=\sum_j\int d\mathbf{r}'\sigma_{ij}(\mathbf{r}\mathbf{r}';\omega)E_j(\mathbf{r}'\omega) \tag{2.3}$$

between the current J and the field, is of the form

$$\sigma_{ij}(\mathbf{r}\mathbf{r}';\omega)=\sigma_{ij}(\omega)\delta(\mathbf{r}-\mathbf{r}').$$

For the general nonlocal conductivity the sum rule states

$$\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\sigma_{ij}'(\mathbf{r}\mathbf{r}';\omega)=\omega_p^2\delta_{ij}\delta(\mathbf{r}-\mathbf{r}'). \tag{2.4}$$

As stated in the Introduction, we shall restrict ourselves to systems which are spatially invariant so that the nonlocal conductivity satisfies

$$\sigma_{ij}(\mathbf{r}\mathbf{r}';\omega)\equiv\sigma_{ij}(\mathbf{r}-\mathbf{r}';\omega)=\int\frac{d\mathbf{k}}{(2\pi)^3}\exp[i\mathbf{k}\cdot(\mathbf{r}-\mathbf{r}')]\sigma_{ij}(\mathbf{k}\omega)$$

and the linear relation (2.3) takes the form

$$J_i(\mathbf{k}\omega)=\sum_j\sigma_{ij}(\mathbf{k}\omega)E_j(\mathbf{k}\omega). \tag{2.3'}$$

The decomposition into transverse (divergence-free) and longitudinal (irrotational) fields then gives

$$\mathbf{J}^T(\mathbf{k}\omega)=\sigma^T(k\omega)\mathbf{E}^T(\mathbf{k}\omega),$$
$$\mathbf{J}^L(\mathbf{k}\omega)=\sigma^L(k\omega)\mathbf{E}^L(\mathbf{k}\omega), \tag{2.3''}$$

with

$$\sigma_{ij}(\mathbf{k}\omega)=\sigma^L(k\omega)(k_ik_j/k^2)+\sigma^T(k\omega)[\delta_{ij}-(k_ik_j/k^2)].$$

The conventionally employed sum rule (2.4) is then written as

$$\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\sigma_{ij}'(\mathbf{k}\omega)=\omega_p^2\delta_{ij}, \tag{2.4'}$$

that is, for all $\mathbf{k}$

$$\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\sigma'^L(k\omega)=\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\sigma'^T(k\omega)=\omega_p^2. \tag{2.4''}$$

The conventional argument for (2.4″) is the following. Equations (2.3″) are equivalent to the statement[5]

$$\mathbf{J}(\mathbf{k},t)=\int_{-\infty}^{\infty}dt'\sigma(k,t-t')\mathbf{E}(\mathbf{k},t')$$

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[2] J. H. Van Vleck, Nuovo Cimento, Suppl. **6**, (1957). Professor Van Vleck has urged the author for many years to point out this error (see pp. 862–863) and give the proper explanation.

[3] M. Tinkham and R. Ferrell, Phys. Rev. Letters **2**, 331 (1959).

[4] See, for example, J. M. Luttinger, Phys. Rev. **135**, A1505 (1964).

[5] We omit the label L or T from the conductivity in these equations, understanding that it may refer to either. We also use the same symbol for the field as a function of frequency and time using the argument and comma to distinguish $\mathbf{E}(k\omega)$ from $\mathbf{E}(k,t)$.

for both transverse and longitudinal fields with

$$\mathbf{J}(\mathbf{k},t)=\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}e^{-i\omega t}\mathbf{J}(\mathbf{k}\omega),\text{ etc.}$$

As long as $\mathbf{E}(\mathbf{k},t)$ vanishes for all $t<0$, its transform $\mathbf{E}(\mathbf{k}z)$ is analytic whenever z is in the upper half-plane. For all such fields, causality requires that $\mathbf{J}(\mathbf{k}z)$ must also be analytic. The ratio $\sigma(kz)=\mathbf{J}(\mathbf{k}z)/\mathbf{E}(\mathbf{k}z)$ is therefore analytic in the neighborhood of each value z_0, at which not all physical fields $\mathbf{E}(\mathbf{k},t)$, which vanish for $t<0$, have $\mathbf{E}(\mathbf{k}z_0)=0$. Since it is possible to exhibit functions $\mathbf{E}(\mathbf{k},t)$ which vanish for $t<0$ and whose transforms do not vanish at any given z_0, it is argued that $\sigma(kz)$ is analytic and consequently that the Kramers-Kronig relations

$$\sigma''(k\omega)=-P\int_{-\infty}^{\infty}\frac{d\omega'}{\pi}\frac{\sigma'(k\omega')}{\omega'-\omega},\tag{2.5a}$$

$$\sigma'(k\omega)=P\int_{-\infty}^{\infty}\frac{d\omega'}{\pi}\frac{\sigma''(k\omega')}{\omega'-\omega},\tag{2.5b}$$

apply.

Furthermore, as $\omega\rightarrow\infty$ any collection of particles behaves as free particles, and for free particles, $\sigma''(k\omega)=\omega_p^2/\omega$. The identities (2.4″) then follow by equating this expression with the high-frequency asymptote of the integral in (2.5a).

It has been pointed out[6] that despite its simplicity the argument is not compelling since the field $\mathbf{E}(k,t)$ or its transform $\mathbf{E}(k\omega)$ is not necessarily a quantity which can be varied arbitrarily. $\mathbf{E}(\mathbf{k},t)$ represents the total microscopic field in the medium: It may be that some fields $\mathbf{E}_0(\mathbf{k},t)$ would give rise to precursor currents according to the relation inferred by analytic continuation from physically attainable fields, but that, in fact, the medium responds in such a way that these *total fields* $\mathbf{E}_0(\mathbf{k},t)$ are impossible to produce. This does not make the ratio of $\mathbf{J}(\mathbf{k}\omega)$ and $\mathbf{E}(\mathbf{k}\omega)$ for physically attainable $\mathbf{E}$ unphysical. It merely implies that for some k the ratio is only measurable (i.e., $\mathbf{E}(\mathbf{k}\omega)$ obtainable) for a range of ω, and that the measureable ratio, together with its continuation, satisfies neither the Kramers–Kronig relation nor the sum rule.

In the subsequent three sections we shall discuss this question more correctly. In Sec. 3, we prove that in a homogeneous system which obeys linear laws the transverse conductivity always satisfies the Kramers–Kronig relation and sum rule. In Sec. 4 we point out that the conventionally defined longitudinal conductivity describes a quantity which may not satisfy these relations for some k by presenting a specific counter-example (the conductivity of a plasma at temperatures slightly above the temperature of a charge density ordering). We argue, however, that as $k\rightarrow 0$, there is no difficulty. In Sec. 5 we point out that this conventional definition is not the one which enters experimental discussions and that a quantity closer to what is experimentally called the conductivity (the ratio of J to E plus the chemical-potential gradient) does satisfy a Kramers–Kronig relation, but a different sum rule.

We employ rationalized Gaussian units, so that, for example, $\omega_p^2=\sum_\alpha n_\alpha e_\alpha^2/m_\alpha$ where n_α, e_α, and m_α are, respectively the density, charge, and mass of the αth species.

3. TRANSVERSE CONDUCTIVITY

Our proof of the Kramers–Kronig relations for the transverse conductivity is based on the dynamical properties (i.e., commutation relations or, classically, Poisson-bracket relations) for the transverse electromagnetic fields. It is precisely because these have no analog for the longitudinal fields that a parallel proof fails in the latter case. We begin by recalling that when we define the transverse conductivity as

$$\mathbf{J}^T(\mathbf{k}\omega)=\sigma^T(k\omega)\mathbf{E}^T(\mathbf{k}\omega)\tag{3.1}$$

and use Maxwell's equations we find

$$[\omega^2-c^2k^2+i\omega\sigma^T(k\omega)]\begin{matrix}\mathbf{E}^T(k\omega)\\ \\ \mathbf{B}^T(k\omega)\end{matrix}=0.\tag{3.2}$$

That is to say, the transverse conductivity is naturally defined in terms of the way the electromagnetic field propagates. Since the field propagation may be induced by an external magnetization and the response to an external disturbance coupled to the magnetic field is described by the retarded commutator of the fields we may define the transverse conductivity in terms of this commutator. As usual, we write[7]

$$\chi_{B_i,B_j}''(\mathbf{k}\omega)=(2\hbar)^{-1}\int_{-\infty}^{\infty}dt\int d\mathbf{r}\times\exp(-i\mathbf{k}\cdot\mathbf{r}+i\omega t)\langle[B_i(\mathbf{r},t),B_j(\mathbf{0},0)]\rangle,$$

$$\chi_{B_iB_j}(\mathbf{k}\omega)=\hbar^{-1}\int_0^{\infty}dt\int d\mathbf{r}\times\exp(-i\mathbf{k}\cdot\mathbf{r}+i\omega t)\langle[B_i(\mathbf{r},t),B_j(\mathbf{0},0)]\rangle$$
$$=\lim_{\epsilon\to 0}\int_{-\infty}^{\infty}\frac{d\omega'}{\pi}\frac{\chi''_{B_iB_j}(\mathbf{k}\omega')}{\omega'-(\omega+i\epsilon)},\tag{3.3}$$

where the brackets indicate an average over a thermal equilibrium density matrix. Because the magnetic field is transverse, $\chi_{B_iB_j}$ and $\chi_{B_iB_j}''$ can be written as

$$\chi_{B_iB_j}(\mathbf{k}\omega)=\chi_{BB}(k\omega)(\delta_{ij}-k_ik_j/k^2).\tag{3.4}$$

The function $\chi_{BB}(k\omega)$ is the boundary value of an

[6] P. Nozières and D. Pines (henceforth NP), *Theory of Quantum Liquids* (W. A. Benjamin, Inc., New York, 1966), Sec. 41.

[7] L. P. Kadanoff and P. C. Martin, Ann. Phys. (N.Y.) **24**, 419 (1963). The notation in NP differs by a minus sign.

analytic function defined as

$$\chi_{BB}(kz)=\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{\chi_{BB}''(k\omega)}{\omega-z}. \tag{3.5}$$

From spatial invariance and time-reversal invariance it follows that $\omega\chi_{BB}''(k\omega)$ is real, even, and positive, so that

$$\chi_{BB}(kz)=\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{\omega\chi_{BB}''(k\omega)}{\omega^2-z^2} \tag{3.6}$$

never vanishes in the upper half-plane. Furthermore, for large z^2

$$\chi_{BB}(kz) \sim -\frac{1}{z^2}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\omega\chi_{BB}''(k\omega)-\frac{1}{z^4}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\omega^3\chi_{BB}''(k\omega). \tag{3.7}$$

Anticipating that

$$\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\omega\chi_{BB}''(k\omega)=c^2k^2,$$

we may define

$$\chi_{BB}(kz)\equiv c^2k^2[c^2k^2-z^2-iz\sigma^T(kz)]^{-1}, \tag{3.8}$$

where $\sigma^T(kz)$ is analytic and approaches zero as $z\rightarrow\infty$. Indeed, we may write

$$\sigma^T(kz)=\int_{-\infty}^{\infty}\frac{d\omega}{\pi i}\frac{\sigma'^T(k\omega)}{\omega-z}=z\int_{-\infty}^{\infty}\frac{d\omega}{\pi i}\frac{\sigma'^T(k\omega)}{\omega^2-z^2}, \tag{3.9}$$

and for large z,

$$\chi_{BB}(kz)\sim-\frac{c^2k^2}{z^2}-\frac{c^2k^2}{z^4}\left(c^2k^2+\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\sigma'^T(k\omega)\right). \tag{3.10}$$

Our proof of the Kramers–Kronig relations and sum rule will therefore be complete if we can show that

$$\begin{aligned}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\omega\chi''_{B_iB_j}(\mathbf{k}\omega) &=\int d\mathbf{r}\exp(-i\mathbf{k}\cdot\mathbf{r})\langle[(i/\hbar)\dot{B}_i(\mathbf{r},0),B_j(\mathbf{0},0)]\rangle\\ &=c^2k^2(\delta_{ij}-k_ik_j/k^2)\end{aligned} \tag{3.11}$$

and

$$\begin{aligned}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\omega^3\chi''_{B_iB_j}(\mathbf{k}\omega) &=\int d\mathbf{r}\exp(-i\mathbf{k}\cdot\mathbf{r})\langle[(i/\hbar)\ddot{B}_i(\mathbf{r},0),\dot{B}_j(\mathbf{0},0)]\rangle\\ &=c^2k^2(c^2k^2+\omega_p^2)(\delta_{ij}-k_ik_j/k^2).\end{aligned} \tag{3.12}$$

Both of these facts follow directly from Maxwell's equations

$$\begin{aligned}\partial\mathbf{B}/\partial t&=-c(\nabla\times\mathbf{E}),\\ \partial^2\mathbf{B}/\partial t^2&=-c(\nabla\times[c(\nabla\times\mathbf{B})-\mathbf{J}]),\end{aligned} \tag{3.13}$$

the definition of the current

$$\begin{aligned}\mathbf{J}(\mathbf{r},t)=\tfrac{1}{2}\sum_\alpha(e_\alpha/m_\alpha)\{\mathbf{p}_\alpha-(e_\alpha/c)\mathbf{A}(\mathbf{r},t),\delta(\mathbf{r}-\mathbf{r}^\alpha(t))\}\\ +c\nabla\times\sum_\alpha g_\alpha\mathbf{s}_\alpha(t)\delta(\mathbf{r}-\mathbf{r}^\alpha(t))),\end{aligned} \tag{3.14}$$

and the commutation relations[8] for the electric and magnetic fields and the vector potential. We have included, since it does not affect the argument, the contribution from magnetic moments, $g_\alpha\mathbf{s}_\alpha(t)$, due to spins.

In the literature it has previously been argued,[9] somewhat incompletely, that the quantity which occurs in the expression for the conductivity is given to order v/c by the transverse current correlation function for a system whose Hamiltonian contains the Coulomb but no magnetic interaction, i.e.,

$$\begin{aligned}\epsilon^T(kz)&=1+i\sigma^T(kz)/z\\ &=1+[\chi_{JJ}{}^{(\text{n.m.i.})T}(kz)-\omega_p{}^2]/z^2.\end{aligned} \tag{3.15}$$

where $\chi_{JJ}{}^{(\text{n.m.i.})T}$ is the rigorous current correlation function for a Hamiltonian with no magnetic terms. [As in the case of the magnetic field correlation function $\chi_{BB}(kz)$, we mean more precisely by $\chi_{JJ}{}^T(kz)$ the continuation of the Fourier transform of the retarded transverse current commutator.] Since all such functions χ have the necessary analytic properties, it follows that $\epsilon^T(kz)$ has them. The proof we have presented eliminates the v/c restriction and the need for filling in the incomplete more complicated arguments which lead to (3.15).[10]

Since the discussions of conductivity[11] are ordinarily carried out in terms of the currents rather than the

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[8] See, for example, P. A. Dirac, *Quantum Mechanics* (Oxford University Press, London, 1947), 3rd ed., Chap. XII.

[9] D. Bohm and D. Pines, Phys. Rev. **82**, 625 (1951). See alternatively NP, Sec. 47, pp. 254–265. Even when fully demonstrated, the expression (2.15) for the conductivity would hold less generally than the authors suggest. Physically, the error would be to order $(v/c)^2$ in the sense that magnetic susceptibilities are of order $(v/c)^2$. The expressions we have given and the proof of the sum rule and dispersion relation apply, for example, in an itinerant ferromagnetic substance, slightly above T_c where it is normal and nonrelativistic but very paramagnetic and where the identity (2.15) does not hold. Our deviation is truly correct to order $(v/c)^2$ in the sense that it is only restricted because we have used a nonrelativistic expression for the particle kinetic energy.

[10] One may of course define, as in NP, $z^2[\epsilon^T(kz)-1]=\chi_{JJ}{}^{\text{sc}T}(kz)-\omega_p{}^2$. Our arguments then show that $\chi_{JJ}{}^{\text{sc}T}(kz)$ is analytic, etc., even when it is not equal to $\chi_{JJ}{}^{(\text{n.m.i})T}(kz)$. We shall reserve the superscript "sc" for a specific combination of correlation functions consistent with the definition in Sec. 6.

[11] These identities are derived and discussed in L. P. Kadanoff and P. C. Martin, Phys. Rev. **124**, 677 (1961). This section provides a concise discussion of the usual approach. [In Eq. (3.31), the alternative to $\alpha^L=\alpha'^L(1-\alpha'^L)^{-1}$ should read $(\epsilon^L)^{-1}-1=1-\epsilon'^L$.] The assumption of the last paragraph of that discussion is the subject of the present investigation. The expressions may also be found in NP, pp. 252–254.

fields, it may be useful to note that by using Maxwell's equations and the field commutation relations we may write

$$\begin{aligned}\epsilon^T(kz)&\\ &=1+i\sigma^T(kz)/z=(c^2k^2/z^2)[1-\chi_{BB}^{-1}(kz)]\\ &=(c^2k^2/z^2)+[1-\chi_{EE}^T(kz)]^{-1}\\ &=(c^2k^2/z^2)+(z^2-c^2k^2)^2[z^2(z^2-c^2k^2+\omega_p^2-\chi_{JJ}^T(kz))]^{-1},\end{aligned} \tag{3.16}$$

where $\chi_{EE}^T(kz)$ and $\chi_{JJ}^T(kz)$ like $\chi_{BB}(kz)$ are the continuations of the Fourier transforms of the retarded electric field and current commutators calculated in the presence of all magnetic interactions. However, in terms of the latter expressions and without the field commutation or Poisson-bracket relations, we have been unable to construct a proof (2.4) and (2.5).

4. LONGITUDINAL CONDUCTIVITY

A. Definitions and Consequences of Analyticity

If the longitudinal conductivity is defined in the conventional manner as the ratio of the measured current to the measured field (or its continuation),

$$\mathbf{J}^L(\mathbf{k}\omega)=\sigma^L(k\omega)\mathbf{E}^L(\mathbf{k}\omega), \tag{4.1}$$

then Maxwell's equations imply

$$[\omega+i\sigma^L(k\omega)]\mathbf{E}^L(\mathbf{k}\omega)=0. \tag{4.2}$$

In this case, similar identities to the transverse ones cited in Sec. 3 apply for the longitudinal conductivity. These are sufficiently familiar[12] so we may cite them without proof[13]:

$$\begin{aligned}\epsilon^L(kz)&\equiv 1+i\sigma^L(kz)/z=[1-\chi_{EE}^L(kz)]^{-1}\\ &=z^2[z^2+\omega_p^2-\chi_{JJ}^L(kz)]^{-1}.\end{aligned} \tag{4.3}$$

In the longitudinal case, the identity of the last two forms incorporates Poisson's equation

$$k^2\chi_{EE}^L(kz)=\chi_{\rho\rho}(kz) \tag{4.4}$$

and the identity

$$z^2\chi_{\rho\rho}(kz)=k^2[\chi_{JJ}^L(kz)-\omega_p^2], \tag{4.5}$$

which follows from current conservation and the commutation relations, and guarantees gauge invariance.[11] Note incidentally that Poisson's equation (4.4) converts the next-to-last identity in (4.3) into perhaps its most familiar form,

$$[\epsilon^L(kz)]^{-1}=1-\chi_{\rho\rho}(kz)/k^2. \tag{4.6}$$

These equations (which imply the analyticity of $[\epsilon^L(kz)]^{-1}$) give the value of $\sigma^L(kz)$ when $\mathbf{E}$ can be produced, and the value of the mathematical continuation when it cannot.

It has been argued somewhat incompletely[14] that under many circumstances the dielectric response may be calculated approximately by treating the long-range interparticle Coulomb potential as an external field to be treated self-consistently but per turbatively, and by taking the remainder to be a short-range potential included in an effective Hamiltonian. If this could be proven, there would be no difficulty in deducing the Kramers–Kronig relations and sum rule. The function

$$\begin{aligned}\epsilon^L(kz)&\equiv 1+[\chi_{JJ}^{\mathrm{sc}L}(kz)-\omega_p^2]/z^2\\ &\equiv 1+\chi_{\rho\rho}^{\mathrm{sc}}(kz)/k^2\end{aligned} \tag{4.7}$$

would be analytic since it would be the response function for an effective Hamiltonian.[7] It would then follow that

$$\epsilon^L(kz)-1=\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{\omega\epsilon^{L''}(k\omega)}{\omega^2-z^2}, \tag{4.8}$$

so that

$$\omega_p^2=\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\sigma^{L'}(k\omega), \tag{4.9}$$

$$\epsilon^L(k0)=1+\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{\epsilon^{L''}(k\omega)}{\omega}, \tag{4.10}$$

In a system with free charge, (4.7) identifies the

[12] J. Lindhard, Kgl. Danske Videnskab Mat.-Fys. Medd. **28,** No. 8 (1954). See also P. Nozieres and D. Pines, Nuovo Cimento **9,** 470 (1958) or NP.

[13] Note that (3.3) and (2.16) are identical when we let $c=0$, since in this limit, the relation between the current and field, whether transverse or longitudinal, is given by $-J=\partial E/\partial t$. Because the combination ck occurs, this is consistent with our expectation that ordinarily, as $k\to 0$, we expect transverse and longitudinal correlations to be identical. The identity also gives a clue to why the long-wavelength and low-frequency limits are not uniform; we can tell whether a disturbance of arbitrarily long wavelength is transverse or longitudinal by waiting longer than the time for a light wave to transverse it, i.e., by making the frequency much less than ck.

[14] For example, S. F. Edwards, Phil. Mag. **3,** 33, 1020 (1958). It is perhaps worth pointing out that in the more rigorous discussions, e.g., of J. S. Langer, Phys. Rev. **127,** 5 (1962) this point is still obscured. Langer has probably correctly studied the irreducible graphs which contribute to the impurity resistance at low concentrations; he has not evaluated what he says he is evaluating—the quantity $\chi_{JJ}''(0\omega)/\omega$ for the Hamiltonian with Coulomb interactions and impurities. One may, of course, also interpret the evaluation although it is not stated as an evaluation of $\chi_{JJ}''^T(0\omega)/\omega$ for the Hamiltonian which neglects magnetic interaction, in line with (2.15). It is then subject to the possible errors discussed in Ref. 9; large corrections due to incipient ferromagnetism, like those due to incipient superconductivity are not easily seen by a Fermi liquid-theory type of analysis. These comments are not intended as an objection to Langer's useful analysis. They are only aimed at clearly separating, to the extent that it is possible, the general discussion of electromagnetic properties without qualifications or approximations from specific (and not so specific) calculational approximations which may be valid at certain temperatures.

inverse screening length k_s^{-1} defined by

$$\epsilon_0 k_s^2 \equiv \lim_{k\to 0} k^2 \epsilon^L(k0) = \lim_{k\to 0} \int_{-\infty}^{\infty} \frac{d\omega}{\pi} \frac{\epsilon''^L(k\omega)k^2}{\omega}, \quad (4.11)$$

$$\epsilon_0 \equiv \lim_{k\to 0} [\epsilon^1(k0) - (\lim_{k\to 0} k^2\epsilon^1(k0))/k^2],$$

with the "compressibility" for the system with screened Coulomb forces. In a two-component system of oppositely charged particles, we may identify

$$\epsilon_0 k_s^2 = \lim_{k\to 0} \int_{-\infty}^{\infty} \frac{d\omega}{\pi} \frac{\chi_{\rho\rho}''^{\mathrm{sc}}(k\omega)}{\omega} = e\left(\frac{\partial\rho}{\partial\mu}\right)_{p,T}. \quad (4.12)$$

(μ is the change in energy with a change in composition keeping the pressure constant and compensating the charge imbalance by an external charge.) If we idealize further to a system of particles moving in a uniform background of positive charge we may write instead

$$\epsilon_0 k_s^2/e^2 = \partial n/\partial\mu \equiv n(\partial n/\partial \bar{p}).$$

The last equality, which is a thermodynamic identity for a one-component system, in this case is nothing more than a definition of the "electronic compressibility." If there were an effective Hamiltonian which described the screened electron interactions, $\bar{p}$ would be the pressure determined by this Hamiltonian. We might alternatively define the electronic compressibility in terms of a $\bar{p}'$ which is given by $\bar{p}' = \bar{p}/\epsilon_0$. It is frequently a good approximation (the random-phase approximation) when the kinetic energy is large compared to the Coulomb energy, to take $\chi_{\rho\rho}^{\mathrm{sc}}/e^2$ equal to the free-particle density correlation function and consequently, to obtain for the inverse square screening radius, classically

$$k_s^2 = e^2(\partial n/\partial\mu) = ne^2\beta$$

and in a degenerate Fermi system

$$k_s^2 = 3ne^2/2\epsilon_F.$$

B. Lack of Analyticity

The question which has been raised concerns under what conditions the effective short-range interactions may be represented by an effective Hamiltonian, the long-range effects being treated classically by a self-consistent equation. If they may, $\epsilon^L(k\omega)$ or $\sigma^L(k\omega)$ can be measured by looking at the ratio of the current and the field. (We recall, however, that since $[\epsilon^L(k\omega)]^{-1}$ describes the result of inelastic electron scattering experiments, there is in principle no difficulty in measuring it by other means.[15]) Mathematically, the necessary and sufficient condition for measurability is that $[\epsilon^L]^{-1}$ has no complex zero. If it has none, ϵ^L will satisfy a dispersion relation and sum rule; if it has one or more, ϵ^L will not.

It is easy to verify that there can be no zero of $[\epsilon^L(kz)]^{-1}$ when $\mathrm{Im}z^2 \neq 0$, and therefore, that the only complex zeros possible occur for $z = iy$. Thus we must show that if $y \neq 0$, then

$$\chi_{\rho\rho}(k, iy) \neq k^2; \qquad \chi_{EE}^L(k, iy) \neq 1;$$
$$\chi_{JJ}^L(k, iy) \neq \omega_p^2 - y^2, \quad (4.13)$$

just as we have shown [Eq. (3.16)] by using the transverse-field commutation relations that in the transverse case in isotropic systems (and therefore implicitly in systems with no magnetic ordering)

$$\chi_{EE}^T(k, iy) \neq 1; \qquad \chi_{JJ}^T(k, iy) \neq \omega_p^2 - y^2 - c^2k^2. \quad (4.14)$$

In the longitudinal case, because $[\epsilon^L(k, iy)]^{-1}$ decreases monotonically as y decreases, there can be at most one value of y for which $\epsilon^L = 0$ or $\chi_{\rho\rho}(k, iy) = k^2$. Nevertheless, since we do not have longitudinal-field commutation relations, we cannot exclude this one possible solution for all k. Indeed, it would appear that it occurs in a system which permits, or almost permits, a static charge density wave solution. In such a system[16] $\chi_{\rho\rho}(k0)$ approaches ∞ for $k \sim 2k_F$ at some transition temperature (perhaps $T = 0$). *At higher temperatures*, before any transition takes place $\chi_{\rho\rho}(2k_F, 0) > (2k_F)^2$ and ϵ is not analytic. We claim that this is approximately the case in a very low density electron gas when it crystallizes. It can be made rigorous by also including nonelectromagnetic long, but not infinite range exchange forces.

C. Heuristic Argument for Longitudinal Sum Rule when $k = 0$

We may obtain some insight into the role of the field commutation relations in our proof of the transverse sum rule by giving the parallel argument that $\sigma^L(0z)$ is analytic in the upper-half complex plane, and hence that

$$\int_{-\infty}^{\infty} \frac{d\omega}{\pi} \sigma^L(0\omega) = \omega_p^2. \quad (4.15)$$

The conclusion is not unexpected, quite apart from the artificial character of the argument. Indeed, if the current correlations have a finite range we expect that $\sigma^L(0z) = \sigma^T(0z)$. This artificial argument leads to the conclusion that $\chi_{\rho\rho}(k^20) \to \infty$ as $k^2 \to 0$. More particularly, it leads to the identity

$$\chi_{EE}^L(00) = \int_{-\infty}^{\infty} \frac{d\omega}{\pi} \frac{\chi_{EE}''^L(0\omega)}{\omega} = 1, \quad (4.16)$$

[15] More precisely, inelastic electron scattering measures charge-density fluctuations as a function of momentum transfer $\hbar k$, and energy loss $\hbar\omega$. The charge fluctuation spectrum is equal to $-\mathrm{Im}[\epsilon^L(k\omega)]^{-1}k^2 = \chi_{\rho\rho}''(k\omega)$ multiplied by $[1 - \exp(-\beta\hbar\omega)]^{-1}$. The connection between the fluctuation spectrum and inelastic scattering has a long history and is frequently attributed to the important paper of L. Van Hove, Phys. Rev. **95**, 249 (1952).

[16] A proof of this physically obvious result is not difficult, but is verbose. We may content ourselves with noting the magnetic analogy. The wave-number-dependent susceptibility diverges for k equal to a reciprocal lattice vector at the Néel temperature.

which we have proven for transverse fields. The argument consists of noting that in the gauge in which ϕ vanishes we may write

$$E_i(\mathbf{r},t)=-(\partial A_i(\mathbf{r},t)/\partial ct),$$

$$\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{\chi_{E_iE_j}''(0\omega)}{\omega}=\int d\mathbf{r}\int_{-\infty}^{\infty}\frac{d\omega}{2\pi}\int_{-\infty}^{\infty}dt \times e^{i\omega t}\langle[A_i(\mathbf{r},t),E_j(\mathbf{0},0)]\rangle\frac{i}{\hbar c}$$

$$=\frac{-i}{\hbar c^2}\int d\mathbf{r}\langle[\dot{A}_i(\mathbf{r},0),A_j(\mathbf{0},0)]\rangle. \tag{4.17}$$

Now if this commutator were valuated in the Lorentz gauge instead of the gauge in which ϕ vanishes and if the covariant commutation relations[17] are supposed to hold in this gauge, then

$$\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{\chi_{E_iE_j}''(0\omega)}{\omega}=\frac{-i}{\hbar c^2}\int d\mathbf{r}\langle[A_i(\mathbf{r},0),\dot{A}_j(0,0)]\rangle=\delta_{ij}. \tag{4.18}$$

The error made in writing $cE_i=-\partial A_i/\partial t$ in the Lorentz gauge comes from the longitudinal terms in $\nabla\phi$ and therefore if the potential correlations are of finite range, the error, which is proportional to k^2, vanishes as $k\to 0$. The distance over which the potential correlations vanish is essentially determined by the distance to which fields penetrate the material. Hence in a perfect dielectric (if there are infinite uniform perfect dielectrics) they are of infinite extent and the argument that $\epsilon^L(k0)\to\infty$ as $k\to 0$ would fail. It is more likely that the conclusion is correct and that any substance which becomes a perfect dielectric undergoes a transition which involves microscopic charge nonuniformity, (the lattice structure is essential), thereby violating the assumption of spatial homogeneity.

5. PHENOMENOLOGICAL DEFINITION OF THE SPATIALLY VARYING CONDUCTIVITY

A. Physical Arguments

Although, as we have stressed, there is no contradiction with causality if the quantity $\sigma^L(kz)$, defined as $\mathbf{J}^L(\mathbf{k}z)/\mathbf{E}^L(\mathbf{k}z)$ or

$$1+i\sigma^L(kz)/z=[1-\chi_{\rho\rho}(kz)/k^2]^{-1}, \tag{5.1}$$

is not analytic, it is a somewhat inelegant state of affairs. We would like to think of the conductivity $\sigma^L(kz)$ not only as a measurable quantity, but also to treat it as we treat other linear response functions. Let us ask, however, whether this expectation is in accord with the experimental situation. It is well known that for finite k, when the charge distribution varies spatially, a compensating chemical potential gradient is set up. Thus with a high density of impurities (or a reduced value for the charge) so that $\omega_p\tau\ll 1$ and $kl\ll 1$ at zero temperature (so that thermoelectric effects are unimportant), the phenomenological equation[18] one would employ is not $J/E=\sigma$ but

$$\mathbf{J}^L=\sigma^L(\mathbf{E}^L-\nabla(\mu/e)). \tag{5.2}$$

This equation implies that the charge variations satisfy an equation of the form

$$-\delta\dot{\rho}=\nabla\cdot\delta\mathbf{J}=\nabla\cdot\delta\mathbf{J}^L=\sigma^L[(\delta\rho/\epsilon_0)+k^2(\partial\mu/\partial\rho e)\delta\rho]$$

or a dispersion relation in plasmas in which the conductivity appears as

$$-\omega=i\sigma[\epsilon_0^{-1}+k^2(\partial\mu/\partial\rho e)]. \tag{5.3}$$

We shall encounter this relation in Sec. 5B. First, however, let us note that in a steady state we have $0=[\epsilon_0^{-1}+k^2(\partial\mu/\partial\rho e)]$, which enables us again to make the identification (4.12)

$$\epsilon_0 k_s^2=\partial\rho e/\partial\mu. \tag{5.4}$$

As exhibited in (5.3), the combination $\sigma(\partial\mu/\partial\rho e)$ plays the role of a diffusion constant D; the ratio of D to the conductivity $\partial\mu/\partial\rho e$ is called the Einstein relation. Actually, it is only a definition, since a given system does not have both a diffusion constant and conductivity in its hydrodynamic description. The relation acquires content when the Coulomb forces contribute negligibly to the diffusion and the screening length is very large. In the simplest approximation discussed earlier, we have, for example, the familiar Einstein relation $D=\sigma/e^2\beta n$.

B. Alternative Definition of Nonlocal Conductivity

If we assume that $\sigma^L(0z)$ is analytic (as argued in Sec. 4) and that we may interchange limits, while we do not obtain (4.9) for arbitrary k, we do have

$$\lim_{k\to 0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\sigma'^L(k\omega)=\omega_p^2 \tag{5.5}$$

and (4.11)

$$\lim_{k\to 0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{\sigma'^L(k\omega)}{\omega^2}=\frac{\epsilon_0 k_s^2}{k^2}.$$

In contrast with

$$1-\chi_{\rho\rho}(kz)/k^2=1-\chi_{EE}^L(kz), \tag{5.6}$$

[17] The extension of the justification of the covariant commutation relations from scattering to these circumstances is not obvious, and this qualification could invalidate our subsequent statements about σ^L as $k\to 0$. We believe the statements are correct, although a better argument would be desirable.

[18] Reference 1, Sec. 25. To conform with the usual notation we have written μ/e, where e is the charge of the lightest species. The quantity $\nabla(\mu/e)$ is well defined when there are many species.

which may vanish for nonvanishing k in some systems, the quantity

$$1-\chi_{EE}{}^L(kz)/\chi_{EE}{}^L(k0)=1-\chi_{\rho\rho}(kz)/\chi_{\rho\rho}(k0) \quad (5.7)$$

[which agrees with (5.6) as $k\to 0$ except in the hypothetical uniform insulator] can never equal zero. We may therefore define $\bar{\sigma}$, which is analytic, by

$$1-\frac{\chi_{\rho\rho}(kz)}{\chi_{\rho\rho}(k0)}\equiv\left[1+\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{\bar{\sigma}'^L(k\omega)}{\omega^2-z^2}\right]^{-1}\equiv\left[1+\frac{i\bar{\sigma}^l(kz)}{z}\right]^{-1}. \quad (5.8)$$

By evaluating both sides asymptotically for large z, we obtain

$$\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\bar{\sigma}'^L(k\omega)=\omega_p{}^2\frac{k^2}{\chi_{\rho\rho}(k0)}=\frac{\omega_p{}^2}{\chi_{EE}{}^L(k0)}. \quad (5.9)$$

The quantity $\bar{\sigma}$ behaves as we expect a transport coefficient to behave.[19] Whenever $\chi_{EE}{}^L(k0)>0$ and there is no ordering (i.e., in any isotropic material), $\bar{\sigma}(k0)$ is positive. It decreases in the neighborhood of a transition in which the charge or field is ordered, i.e., where $\chi_{EE}{}^L\to\infty$. The point at which a pole appears in ϵ^L or σ^L, i.e., at which $\chi_{\rho\rho}(k0)/k^2=\chi_{EE}{}^L(k0)=1$ as contrasted with infinity, has no significance other than indicating that $\sigma^L(kz)$ is an artificial quantity.

To the extent that $\bar{\sigma}/\sigma$ may be taken to be frequency-independent, we may also infer from (5.9) in conformity with (5.3) that

$$\bar{\sigma}^L(k0)\cong\sigma k^2/\chi_{\rho\rho}(k0)=\sigma[1+(\partial\mu/\partial\rho e)k^2]. \quad (5.10)$$

With this replacement, at long wavelengths and low frequencies, where the dispersion relation for charge fluctuations

$$\omega+i\bar{\sigma}^L(k\omega)=0 \quad (5.11)$$

reduces (for $\epsilon_0=1$) to the phenomenological equation

$$\omega+i\sigma[1+k^2(\partial\mu/\partial e\rho)]=0,$$

$\bar{\sigma}^1$ is k-independent. Thus the generally more appropriate conductivity function $\bar{\sigma}^1$ is closer to the phennomenological description, and coincides with it in this suitably simplified example.

At finite temperatures, there will be electric conduction and thermal conduction modes. For arbitrary k, both modes will appear as solutions to $\omega+i\bar{\sigma}^L(k\omega)=0$. For this reason one may question whether even $\bar{\sigma}(k\omega)$ should be called a nonlocal conductivity. The situation is similar to the one which exists for momentum fluctuations in a fluid.[20] There, because the transverse modes are uncoupled at all temperatures, it is possible to define a complex frequency and wave-number–dependent shear viscosity or shear modulus. However, because the longitudinal sound mode is coupled with thermal conduction at finite temperatures, the coefficient one might be tempted to call the wave-number–dependent longitudinal viscosity contains also the effects of thermal conduction on the velocity and damping of the sound wave mode and gives the solution to the dispersion equation which corresponds to the thermal conduction mode. In the electrical case this coupling is weaker and vanishes at $k\to 0$.

[19] The argument that the analogous quantity in a neutral system (the diffusion constant) vanishes is a general one. The diffusion constant is given by a transport coefficient, which is relatively regular, divided by an infinite susceptibility. Experimental verification of this slowing down at the critical temperature has been the subject of several recent investigations.

[20] L. P. Kadanoff and P. C. Martin, Ann. Phys. (N.Y.) **24**, 454 (1963).

6. TRANSPORT COEFFICIENT IN CHARGED SYSTEMS

In the previous sections we have investigated in some detail the nonlocal conductivity. A special case, of course, is the local conductivity, the transport coefficient which occurs in Ohm's law. We may write it alternatively as

$$\begin{aligned}\sigma'&=\lim_{\omega\to 0}\lim_{k\to 0}\sigma'(k\omega)\\&=\lim_{\omega\to 0}\lim_{k\to 0}|\epsilon^L(k\omega)|^2\chi_{JJ}{}''^L(k\omega)/\omega\\&=\lim_{\omega\to 0}\lim_{k\to 0}\chi_{JJ}{}''^{L\mathrm{sc}}(k\omega)/\omega\\&=\lim_{\omega\to 0}\lim_{k\to 0}\omega\chi_{\rho\rho}{}''^{\mathrm{sc}}(k\omega)/k^2.\end{aligned} \quad (6.1)$$

As noted here and stressed previously elsewhere,[21] it is not, as is frequently stated, the same as the diffusion constant in a neutral system,

$$\begin{aligned}(\partial n/\partial\mu)D&=\lim_{\omega\to 0}\lim_{k\to 0}\chi_{JJ}{}''^L(k\omega)/\omega\\&=\lim_{\omega\to 0}\lim_{k\to 0}\omega\chi_{\rho\rho}{}''(k\omega)/k^2,\end{aligned} \quad (6.2)$$

because of the long-range Coulomb forces. Indeed, because of the Coulomb forces, the thermodynamics of a charged system are really simpler than those of a neutral system. For simplicity, and to conform with earlier discussions of this problem, we shall discuss the case in which there is a single charge carrier. The compensating background is taken to be inert as are the infinitely heavy neutral impurities we shall suppose produce the resistance. In this case the only thermodynamic variables are the ones which occur in a one-component neutral system with infinitely heavy impurities—total mass, total momentum, and total energy. The concentration is not a thermodynamic variable which can be varied quasistatically because the long-range Coulomb forces restore neutrality in a finite time of order $\omega_p{}^{-1}$. As might be expected, this simplifies the hydrodynamic description and consequently the expressions for the transport coefficients.

[21] P. C. Martin, in *Statistical Mechanics of Equilibrium and Non-Equilibrium*, edited by J. Meixner (North-Holland Publishing Company, Amsterdam, 1965).

Indeed, just as in a one-component system we may prove that the thermal conductivity κ is given by

$$\kappa T=\lim_{\omega\to 0}\lim_{k\to 0}\omega\chi_{\epsilon\epsilon}''(k\omega)/k^2, \tag{6.3}$$

where ϵ is the energy density operator. Because the charge gradients are not maintained on a hydrodynamic time scale, we have (as in a one-component system)

$$\lim_{\omega\to 0}\lim_{k\to 0}\omega\chi_{\epsilon\rho}''(k\omega)/k^2=0,\quad \lim_{\omega\to 0}\lim_{k\to 0}\omega\chi_{\rho\rho}''(k\omega)/k^2=0 \tag{6.4}$$

and consequently in a rigorous evaluation we have for any λ,

$$\kappa T=\lim_{\omega\to 0}\lim_{k\to 0}\chi_{\epsilon-\lambda\rho,\epsilon-\lambda\rho}''(k\omega)/\omega. \tag{6.5}$$

In other words, the expression in terms of a commutator (for the thermal conductivity) in a two-component charged system is identical with the expression in a neutral one-component system.[21]

The situation is also quite simple for the thermoelectric power. In this case it is convenient to take as the defining experiment the one which can be characterized by an open circuit (i.e., with a closed system).

Since such a system is characterized by a constant chemical potential, we have, when we adiabatically apply a force which can be shown to correspond to a change in temperature and charge density in the long-wavelength limit,

$$\delta\rho=T(\partial\rho/\partial T)\delta T/T,$$
$$\delta\rho=\chi_{\rho,\epsilon-\mu\rho/e}(\delta T/T)\equiv\chi_{\rho,q}(\delta T/T), \tag{6.6}$$

whence, in this limit

$$\delta\mathbf{E}=(\chi_{\rho,q}/k^2)(\nabla T/T). \tag{6.7}$$

The quantity $\chi_{\rho,q}/k^2$ has a limit as k and ω approach zero, which is independent of order (just as $\chi_{\rho\rho}/k^2\to 1$ as k and ω approach zero independent of the order). Were we to describe this same ratio between field and temperature in terms of the usual phenomenological equations, we would write

$$\mathbf{E}=(\pi/T)\nabla T=(K_{12}/\sigma T)\nabla T, \tag{6.8}$$

where K_{12} is the conventional Onsager coefficient,[22]

$$\mathbf{J}=\sigma(\mathbf{E}-\nabla(\mu/e))-(K_{12}/T)\nabla T,$$
$$\mathbf{J}^q=K_{21}e(\mathbf{E}-\nabla(\mu/e))-(\kappa+K_{12}K_{21}/\sigma T)\nabla T. \tag{6.9}$$

Thus,

$$K_{12}/\sigma=\lim_{k\to 0}\lim_{\omega\to 0}\chi_{\rho,q}/k^2=\lim_{\omega\to 0}\lim_{k\to 0}\chi_{\rho,q}/k^2. \tag{6.10}$$

These definitions appear to be very different from the ones which are usually employed. However, we may connect them with the ordinarily applied ones by the following identities[23]:

$$\chi_{AB}^{\mathrm{sc}}(k\omega)\equiv\chi_{AB}(k\omega)-\chi_{A\rho}(k\omega)\chi_{\rho B}(k\omega)[\epsilon^L(k\omega)/k^2],$$
$$\chi_{AB}(k\omega)\equiv\chi_{AB}^{\mathrm{sc}}(k\omega)-\chi_{A\rho}^{\mathrm{sc}}(k\omega)\chi_{\rho B}^{\mathrm{sc}}(k\omega)[k^2\epsilon^L(k\omega)]^{-1}. \tag{6.11}$$

These identities have a significance in perturbation theory, and are in accord with our earlier definition of $\chi_{\rho\rho}^{\mathrm{sc}}(k\omega)$. In general, $\chi_{AB}^{\mathrm{sc}}(k\omega)$ defined by these equations is not a correlation function [and as we have argued earlier for $\chi_{\rho\rho}^{\mathrm{sc}}(k\omega)$, need not have the analytic properties of a correlation function]. Nonetheless, since

$$\lim_{\omega\to 0}\lim_{k\to 0}\omega\epsilon^L(k\omega)=i\sigma^L,$$

we may write

$$K_{12}=\lim_{\omega\to 0}\lim_{k\to 0}\chi_{\rho,q}(k\omega)\omega\epsilon^L(k\omega)/ik^2 \tag{6.12}$$

$$=\lim_{\omega\to 0}\lim_{k\to 0}\chi_{\rho,q}^{\mathrm{sc}}(k\omega)\omega/ik^2. \tag{6.13}$$

Even in superconductors the imaginary part of this term vanishes, so that

$$K_{12}=\lim_{\omega\to 0}\lim_{k\to 0}\chi_{\rho,q}''^{\mathrm{sc}}(k\omega)\omega/k^2$$
$$=\lim_{\omega\to 0}\lim_{k\to 0}\chi_{JJ^q}''^{\mathrm{sc}}(k\omega)/\omega. \tag{6.14}$$

(The last equality involves a few manipulations since the screened functions are not correlation functions but combinations of them.) Furthermore, we may write

$$\sigma'^L=\lim_{\omega\to 0}\lim_{k\to 0}\chi_{\rho\rho}''^{\mathrm{sc}}(k\omega)\omega/k^2. \tag{6.15}$$

Finally, if we express the thermal conductivity in terms of screened quantities, we find

$$\frac{\omega\chi_{qq}(k\omega)}{k^2}=\frac{\omega}{k^2}\chi_{qq}^{\mathrm{sc}}-\frac{\omega^2}{k^2}\frac{(\chi_{q\rho}^{\mathrm{sc}})^2}{k^2(\omega+i\sigma^L(k\omega))}. \tag{6.16}$$

Therefore,

$$\kappa T=\lim_{\omega\to 0}\lim_{k\to 0}\omega\chi_{qq}''(k\omega)/k^2$$
$$=\lim_{\omega\to 0}\lim_{k\to 0}\chi_{J^qJ^q}''(k\omega)/\omega \tag{6.17}$$

may also be written as

$$\kappa T=\lim_{\omega\to 0}\lim_{k\to 0}\left[\frac{\chi_{J^qJ^q}''^{\mathrm{sc}}(k\omega)}{\omega}-\frac{K_{12}^2\sigma'}{(\sigma')^2+(\sigma'')^2}\right]. \tag{6.18}$$

Since σ'' is proportional to ρ_s/ω, the last term vanishes

[22] J. M. Luttinger, Phys. Rev. **136**, A1481 (1964).

[23] This identity is related to the elimination of the Coulomb force in terms of the screened dynamic Coulomb force which one carries out diagrammatically or in terms of functional derivatives. The dynamic force cannot be described in terms of a Hamiltonian.

in a superconductor and we may write[22]

$$\kappa T = \lim_{\omega\to 0}\lim_{k\to 0} \chi_{J^qJ^q}{}''^{\mathrm{sc}}(k\omega)/\omega = \lim_{\omega\to 0}\lim_{k\to 0} \chi_{J^qJ^q}{}''(k\omega)/\omega. \tag{6.19}$$

The first equality which is restricted to superconductors is not a Kubo-like expression in terms of a correlation function; the second, which is a Kubo-like expression, is not restricted to superconductors.

Clearly, as the charge is turned off we obtain the equations we have noted elsewhere[21] for neutral systems;

$$\mathbf{J}/e = -D(\partial n/\partial\mu)\nabla\mu - L_{12}(\nabla T/T),$$

$$\mathbf{J}^q = -L_{12}\nabla\mu - (\kappa + L_{12}{}^2(\partial n/\partial\mu)/DT)\nabla T, \tag{6.20}$$

the combinations

$$\lim_{\omega\to 0}\lim_{k\to 0}\lim_{e\to 0} \chi_{JJ}{}''^{\mathrm{sc}}(k\omega)/\omega e^2 \to D(\partial n/\partial\mu)_T,$$

$$\lim_{\omega\to 0}\lim_{k\to 0}\lim_{e\to 0} \chi_{J^qJ}{}''^{\mathrm{sc}}(k\omega)/\omega e \to L_{12},$$

$$\lim_{\omega\to 0}\lim_{k\to 0}\lim_{e\to 0} \chi_{J^qJ^q}{}''^{\mathrm{sc}}(k\omega)/\omega \to \kappa T + L_{12}{}^2(\partial\mu/\partial n)/D, \tag{6.21}$$

approaching the rigorous Kubo-like expressions for neutral systems. When the neutral limit $e\to 0$ is taken first, an additional hydrodynamic mode develops and the thermodynamic description is modified because the concentration becomes a thermodynamic variable as $\omega_p\to 0$. In other words, the nonhydrodynamic plasma mode becomes the hydrodynamic diffusion mode. Provided that the screening length is much larger than any other characteristic length, we have two domains in the charged system: $k < k_s$, where there are Coulomb-like phenomena and no hydrodynamic diffusion mode, and $k_s < k < 1/l$, where the Coulomb effects are unimportant and we may speak of a diffusion constant. The conductivity in the former wavelength domain and the approximate diffusion constant in the latter are connected by the Einstein relation. Equivalently, we may say that the plasma mode which behaves as $\omega^2 \cong \omega_p{}^2 + iDk^2\omega_p$ when $k \ll k_s$ behaves as a diffusive mode $\omega \cong iDk^2$ when $Dk^2 \gg \omega_p$ and $k \gg k_s$.

It might be true that if magnetic effects are neglected the arguments which state

$$\chi_{JJ}{}^{\mathrm{sc}L}(k\omega) \cong \chi_{JJ}{}^{(\mathrm{n.m.i})T}(k\omega) \tag{6.22}$$

also apply to the energy current. If this is truly the case, it would be possible to take the definitions

$$K_{12} = \lim_{\omega\to 0}\lim_{k\to 0} \chi_{J^qJ}{}^{(\mathrm{n.m.i})''T}(k\omega)/\omega, \tag{6.23}$$

$$\kappa T + K_{12}{}^2/\sigma = \lim_{\omega\to 0}\lim_{k\to 0} \chi_{J^qJ^q}{}^{(\mathrm{n.m.i})''T}(k\omega)/\omega. \tag{6.24}$$

In addition to being artificial, such a procedure[24] is at best only approximate and requires proof. It also requires some examination of the relevant current operators. We have been careful to avoid discussing either the longitudinal or transverse energy current because when long-range forces are present, explicit examination shows that they are peculiar surface-dependent operators for which it is unclear that limits exist. The energy density, itself, even though it is less singular, is a sensitive extremely nonlocal operator when there are Coulomb forces.[25]

We may summarize this section as follows: (1) The simplest current correlation expression for the thermal conductivity is always rigorously correct (it is not correct only in superconductors); (2) the usually written current correlation function expression for the thermopower, like the Kubo expression for the electric conductivity, is never correct; (3) the usual expressions for the thermal conductivity, thermopower, and conductivity are correct when expressed in terms of "screened" response functions, but screened response functions are not correlation functions of currents for an effective Hamiltonian, and need not have the analytic properties of correlation functions, i.e., there is no H^{sc} such that one can write a Kubo formula $\chi_{AB}{}''^{\mathrm{sc}} = (\mathrm{Tr}[A,B]\exp(-\beta H^{\mathrm{sc}}))/\mathrm{Tr}\exp(-\beta H^{\mathrm{sc}})$; (4) these statements all come about because of the long-range Coulomb force, and consequently when $e\to 0$, the usually quoted expressions become rigorously correct.

7. SUMMARY

In this section we tabulate the results we have derived, together with other properties of the current correlation functions. In isotropic materials, in addition to the well-known result for all k,

$$-\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\omega\,\mathrm{Im}\left[\frac{1}{\epsilon^L(k\omega)}\right] = \int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{\chi_{JJ}{}''^L(k\omega)}{\omega} = \omega_p{}^2, \tag{7.1}$$

we have defined (5.8) a longitudinal conductivity $\bar{\sigma}^L(k\omega)$ which for all k, satisfies the Kramers–Kronig

[24] Under these implicit assumptions, Ref. 4 is probably correct except for magnetic (not v/c) corrections.

[25] Although we have not investigated the question in detail, the rigorous formula which relates the thermopower to the conductivity in terms of a thermodynamic derivative suggests that it may be possible, as Lord Kelvin originally argued, to obtain the reciprocity relation without the irreversible thermodynamics which is necessary for neutral systems.

relations and[26]

$$\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\bar{\sigma}'^L(k\omega)=\omega_p^2k^2\chi_{\rho\rho}^{-1}(k0)=\omega_p^2\left[\frac{\epsilon^L(k0)}{\epsilon^L(k0)-1}\right]\geq 0. \quad (7.2)$$

In a perfect insulator we can only write

$$\lim_{k\to 0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\bar{\sigma}'^L(k\omega)=\omega_p^2\left(\frac{\epsilon}{\epsilon-1}\right)\geq 0 \quad (7.3)$$

when there is free charge $\epsilon\to\infty$ as $k\to 0$, so that

$$\lim_{k\to 0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\bar{\sigma}'^L(k\omega)=\omega_p^2. \quad (7.4)$$

Both in a conductor where $\chi_{\rho\rho}(k0)\to k^2$ as $k\to 0$, and in an idealized uniform insulator, when (7.4) need not be true but where we may suppose that $\sigma^L(k\omega)=\sigma^T(k\omega)$ as k and ω approach zero, we may write for the conventionally defined longitudinal conductivity,

$$\lim_{k\to 0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\sigma'^L(k\omega)=\omega_p^2. \quad (7.5)$$

Besides (7.3)–(7.5), we have the usual long-wavelength sum rules. First, the analog of the compressibility sum rule for a neutral system[27]

$$\lim_{k\to 0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\frac{\chi_{nn}''(k\omega)}{\omega}=n\left(\frac{\partial n}{\partial p}\right)_T,$$

namely,

$$\lim_{k\to 0}-k^2\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{1}{\omega}\,\mathrm{Im}\left[\frac{1}{\epsilon^L(k\omega)}\right]$$

$$=\lim_{k\to 0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\frac{\chi_{\rho\rho}''(k\omega)}{\omega}=\left(\frac{\partial\rho}{\partial\phi}\right)_T=0 \quad (7.6)$$

[26] Equation (7.2) is the analog of the sum rule for the damping constant for transverse oscillations of an uncharged fluid. There the quantity which gives the dispersion relation,

$$\omega^2+iD^T(k\omega)k^2\omega=0,$$

satisfies

$$\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,D^{T\prime}(k\omega)=\frac{c_\infty^2(k)}{\chi_{gg}^T(k0)}.$$

If the fluid is a glass, i.e., if it supports transverse waves at long wavelengths then the constant $D^{T\prime}$ contains a δ function, i.e.,

$$D^{T\prime}(k\omega)=c_0^2(k)\delta(\omega)/\chi^T(k0)+D^{T\prime\,\mathrm{reg}}(k\omega).$$

The analogous δ-function term in the conductivity is associated with superconductivity in the electrical case. There is a further analogy. In the normal fluid $\chi_{gg}^T(k0)\to mn$ as $k\to 0$. Likewise, $\chi_{\rho\rho}(k0)/k^2=\chi_{EE}(k0)\to 1$ in a conductor.

[27] See, for example, the discussion of thermodynamic sum rules in L. P. Kadanoff and P. C. Martin, Ann. Phys. (N.Y.) **24**, 450 (1963). The compressibility sum rul itself has been known for many years.

and the stronger statement

$$\lim_{k\to 0}-\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{1}{\omega}\,\mathrm{Im}\left[\frac{1}{\epsilon^L(k\omega)}\right]$$

$$=\lim_{k\to 0}\left(1-\frac{1}{\epsilon^L(k\omega)}\right)$$

$$=\lim_{k\to 0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\frac{\chi_{EE}''^L(k\omega)}{\omega}=1. \quad (7.7)$$

In this stronger form, the coefficient of k^2 defines a screening radius $(\epsilon_L^{-1}=k^2/k_s^2\epsilon_0)$. In a system with a single carrier this quantity may be related to the local chemical potential $\epsilon_0 d\mu\equiv ed\rho k_s^{-2}$. The diffusion constant D, defined as $\bar{\sigma}=\sigma=Dk_s^2\epsilon_0$, will be related to measurable diffusion if there is a hydrodynamic description applicable for $k_s\ll k\ll l^{-1}$, where l is a mean free path.

Apart from the idealized uniform perfect insulator where we can only write

$$\epsilon^L(00)-1=\lim_{k\to 0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{1}{\omega}\,\mathrm{Im}\left[\frac{1}{1-\chi_{EE}^L(k\omega)}\right]$$

$$=\lim_{k\to 0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{1}{\omega}\,\mathrm{Im}\epsilon^L(k0)\geq 0, \quad (7.8)$$

we have

$$\epsilon_0k_s^2=\lim_{k\to 0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{k^2}{\omega}\,\mathrm{Im}\left[\frac{1}{1-\chi_{EE}^L(k\omega)}\right]$$

$$=\lim_{k\to 0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{k^2}{\omega}\,\mathrm{Im}\epsilon^L(k\omega)\geq 0. \quad (7.9)$$

The equations (7.2)–(7.5) and (7.8) and (7.9) are the analogs, respectively, of the two statements

$$\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\omega\,\mathrm{Im}\epsilon^L(k\omega)=\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\sigma'^L(k\omega)=\omega_p^2 \quad (7.10)$$

and

$$\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\mathrm{Im}\left[\frac{\epsilon^L(k\omega)}{\omega}\right]=\epsilon'^L(k0)-1, \quad (7.11)$$

which are true for all k when $\epsilon^L(kz)$ is analytic but which can be violated for $k\neq 0$ in a causal linear homogeneous medium far from a critical point. We may rationalize the insignificance of their failure for $k\neq 0$ by noting that even to order k^2, it is not σ^L but $\bar{\sigma}^L$ which is the appropriate phenomenological coefficient.

On the contrary, for transverse fields we always have the analog of (7.10),

$$\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\omega\,\mathrm{Im}\epsilon^T(k\omega)=\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\sigma'^T(k\omega)=\omega_p^2, \quad (7.12)$$

and of (7.11),

$$P\int_{-\infty}^{\infty}\frac{d\omega'}{\pi}\frac{\operatorname{Im}\epsilon^T(k\omega')}{\omega'-\omega}=\operatorname{Re}[\epsilon^T(k\omega)-1], \quad (7.13)$$

even though we can not,[28] in general, deduce the analog of (7.1), i.e.,

$$\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{\chi_{JJ}''^T(k\omega)}{\omega}\neq\omega_p^2 \quad (7.14)$$

for arbitrary k, nor even for $k=0$, in a superconductor. To deduce (7.14) for $k\to0$, we must argue that the current correlations do not extend to infinity, (which is true except in superconductors[29]), so that as $k\to0$, $\chi_{JJ}''^T(k\omega)\to\chi_{JJ}''^L(k\omega)$ and employ (7.1).

To make contact with other statements about the transverse conductivity—including a few prevalent ones about the transverse conductivity in superconductors that our conclusions might seem to contradict—we note that the statement for magnetic properties which corresponds to (7.8) is

$$\frac{1}{\mu}=\lim_{k\to0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{1}{\omega}\operatorname{Im}\left[\frac{1}{\chi_{BB}(k\omega)}\right]=\left(\frac{\partial B}{\partial H}\right)^{-1}$$

$$\lim_{k\to0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{\omega^2}{c^2k^2}\operatorname{Im}\left[\frac{1}{1-\chi_{EE}^T(k\omega)}\right], \quad (7.15)$$

and in a superconductor the analog of (7.9) is[30]

$$\frac{k_L^2}{\mu_0}=\lim_{k\to0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{k^2}{\omega}\operatorname{Im}\left[\frac{1}{\chi_{BB}(k\omega)}\right]$$

$$=\lim_{k\to0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{\omega}{c^2}\operatorname{Im}\left[\frac{1}{1-\chi_{EE}^T(k\omega)}\right]. \quad (7.16)$$

This latter equation defines the magnetic screening length, or equivalently, the number of superconducting electrons $c^2k_L^2=(n_s/n)\omega_p^2\mu_0$.

As in the electrical case, the more completely meaningful quantity is the static nonlocal permeability $\chi_{BB}(k0)=\mu(k)$, which plays a role parallel to the static nonlocal dielectric constant $\epsilon^L(k0)$. Its singularities, rather than its behavior for small k, may determine the field at large distances.[31]

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[28] This point has been discussed at considerable length in superconductivity where the discrepancy is greatest. The first to stress the inequality in the literature was P. W. Anderson, Phys. Rev. **110**, 827 (1958).

[29] M. R. Schafroth, Phys. Rev. **100**, 502 (1955). The argument, which states more precisely that the volume integral of the current correlation function is finite, is probably correct except in superconductors.

[30] Over the years a great deal has been written about the relationship between an energy gap, on the one hand, and the Meissner effect and superconductivity on the other hand. While the Meissner effect certainly does not require an energy gap, the converse is true in a sense. If the frequency integral of the measured absorption does not exhaust the sum rule (it does in insulators) for values of k for which the dispersion is anomalous, there is a Meissner effect.

The objections to the argument that the Meissner effect and superconductivity are equivalent also seem superfluous to us. A superconductor is characterized by the fact that in isolation it can support a spatially varying time-independent current (superconductivity) and magnetic field (the compensating induced magnetic field which gives rise to the Meissner effect) although (in contrast with a ferromagnet) its equilibrium state has no magnetization or current. The two statements are equivalent since time-independent spatially varying magnetic fields imply [by the microscopic Maxwell equation $\nabla\times B=(1/c)J$] time-independent currents, and conversely. The dispersion-relation and sum-rule argument is a restatement of this fact.

To choose as counterexample to the above argument about their equivalence a free-electron gas in a uniform background of positive charge is to becloud the issue since the counterexample, in addition to being unphysical, is a system for which the conductivity is not infinite but nonexistent. It is true that a long-wavelength plasma oscillation will persist forever, but that is not how one measures the conductivity. Furthermore, we do not want to call any system in which a nonuniform current propagates for an infinitely long time a superconductor. By that definition an insulator, which has persistent induced diamagnetic currents associated with the persistent propagation of electromagnetic waves, would be a superconductor. (Mathematically, the insulator has a pole for ω real and $\omega\neq0$, the superconductor, a pole at $\omega=0$; both lead to nondecaying waves.)

One can produce a mathematical definition for the conductivity

$$\lim_{\omega\to0}\lim_{k\to0}\sigma(k\omega),$$

and this limiting process does give a vanishing conductivity to the insulator and an infinite conductivity to free particles, but to relate this mathematical definition to any measurement requires: (1) the existence of a mean free path l, (2) the argument that the limit $k\to0$ may be freely taken whenever the wave number of the field k, satisfies $kl\ll1$. The measurement involves a steady state with no accelerating currents and with a spatially varying magnetic field. Such a state cannot be produced in a free-electron gas, and so the mathematical limit has nothing to do with a measurement in this case. An equally confusing and similar statement would be that a neutral one-component system has an infinite diffusion constant; in fact the diffusion constant is undefined and unmeasurable.

For this reason we believe that the discussion of G. Rickayzen and W. A. B. Evans, Ann. Phys. (N.Y.) **33**, 275 (1965), which demonstrates that the mean free path has the same effect on the conductivity of the normal component of a superconductor as it has on the conductivity of a normal metal, is quite unnecessary for answering the question of the equivalence of superconductivity and Meissner effect—a question which assumes that we are talking about systems with defined measurable conductivities.

[31] It should be apparent that the combinations $\epsilon_0k_s^2=e\partial\rho/\partial\mu$ and $k_L^2/\mu_0=(n_s/n)(\omega_p^2/c^2)$ play a more fundamental role than the individual terms k_s^2 and ϵ_0 or k_L^2 and μ_0^{-1}. Their independent definitions are based on the small wave-number expansion of $\epsilon(k0)$ and $\mu(k)$. Under some circumstances this small wave-number expansion is applicable for determining the field penetration depth. The Debye screening of a classical plasma and the magnetic screening of a superconductor for which the London equations apply are two examples. On the other hand there are several circumstances in which the small wave-number expansion does not determine the dominant long wavelength dependence or is inapplicable at the penetration depth it determines. One example is the Friedel oscillations which arise from the singularity in $\epsilon(k0)$ at $k=2k_F$; another is the typical weak coupling type-I superconductor whose coherence length ζ is much larger than k_L^{-1}, and whose magnetic penetration length lies between the two.

Like the electric field penetration length argument, the magnetic penetration length argument can be expressed in simple terms. The London equation is: $c\nabla\times J=(n_s/n)\omega_p^2B$ and Maxwell's equation is $c\nabla\times H=c\nabla\times B/\mu_0=J$, when we use the $k=0$ values for the coefficients $(n_s/n)\omega_p^2$ and μ_0^{-1}. The conclusion, $k_Z^2=(n_s/n)(\omega_p^2/c^2)\mu_0$, is therefore limited to the case in which their wave-number dependence may be neglected.

The quantity $\mu(k)$ is related to the transverse conductivity in the following way: In general we have[32]

$$\sigma'^T(k\omega)=c^2k^2\pi\delta(\omega)[-1+\mu^{-1}(k)]+\sigma'^{\text{reg }T}(k\omega),$$
$$\sigma^T(kz)=-(ic^2k^2/z)[1-\mu^{-1}(k)]+\sigma^{\text{reg }T}(kz). \quad (7.17)$$

The sum rule (7.12) may therefore be written for $\sigma^{\text{reg }T}$ as

$$\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\sigma'^{\text{reg }T}(k\omega)=\omega_p^2-c^2k^2[\mu^{-1}(k)-1], \quad (7.18)$$

which implies that[33]

$$\mu^{-1}-1\leq\omega_p^2/c^2k^2; \qquad \mu\geq(1+\omega_p^2/c^2k^2)^{-1}. \quad (7.19)$$

Except in superconductors we have

$$\lim_{k\to0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\sigma'^{\text{reg }T}(k\omega)=\lim_{k\to0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\sigma'^T(k\omega)=\omega_p^2. \quad (7.20)$$

These equations provide in a somewhat concise fashion the resolution of the paradox concerning diamagnetism which we posed in the Introduction. To spell it out more comprehensibly, that paradox is the result of a particular separation of the effects of an external transverse field into magnetic and electric parts. All of the effects are described in terms of σ^T or $\chi_{JJ}{}^T$, and $\sigma^{T\prime}$ and $\chi_{JJ}{}^{T\prime\prime}/\omega$ must be positive definite. However, the conventional description entails writing

$$\begin{aligned}J_i&=\sigma^TE_i^T+\sigma^LE_i^L\\&=\sigma^LE_i+(\sigma^T-\sigma^L)E_i^T\\&=\sigma^LE_i+(\sigma^T-\sigma^L)(i\omega/ck^2)(\nabla\times B)_i\\&=\sigma^LE_i+c(\nabla\times M)_i,\end{aligned}$$

whence

$$\begin{aligned}i\omega(\sigma^T-\sigma^L)/c^2k^2&=1-\mu^{-1}\equiv\chi_{MM}/(1+\chi_{MM})\\&=(\omega^2/c^2k^2)(\epsilon^T-\epsilon^L).\end{aligned}$$

For simplicity, suppose magnetic effects are small. Then these quantities are the same as

$$\begin{aligned}\chi_{MM}(\omega)&\cong(\chi_{JJ}{}^{T(\text{n.m.i})}-\chi_{JJ}{}^{L(\text{n.m.i})})/c^2k^2\\&=\lim_{k\to0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{\chi^{T\prime\prime}(k\omega)-\chi^{L\prime\prime}(k\omega)}{\omega c^2k^2}.\end{aligned}$$

The quantity $[\chi''^T(\omega)-\chi''^L(\omega)]/k^2$ represents the total work done by the external magnetic field minus the work which we have chosen to describe in terms of its associated electric field. There is no reason why the difference need be positive. Indeed, the orbital contribution to the difference is usually negative (i.e., diamagnetic). The proper deduction is just the one contained in (7.19), or equivalently,

$$\chi_{MM}(0)>-\omega_p^2/[\omega_p^2+c^2k^2],$$

and, as we might expect, as $k\to0$ this is precisely the familiar thermodynamic restriction $\chi_{MM}>-1$ or $\mu>0$.

In superconductors, as $k\to0$

$$\mu^{-1}(k)-1\to(\omega_p^2/c^2k^2)(n_s/n) \quad (7.21)$$

and so

$$\lim_{k\to0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\,\sigma'^{\text{reg }T}(k\omega)=\omega_p^2\,\frac{n_n}{n}. \quad (7.22)$$

This equation is the one referred to when it is stated that the transverse sum rule is violated. Substituting the relations between $\sigma^{\text{reg }T}$ and $\chi_{JJ}{}^T$, and between $\sigma^{\text{reg }T}$ and $\chi_{JJ}{}^{(\text{n.m.i})T}$, we see alternatively that

$$\lim_{k\to0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{\chi_{JJ}{}''^{(\text{n.m.i})}(k\omega)}{\omega}=\lim_{k\to0}\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{\chi_{JJ}{}''^T(k\omega)}{\omega}=\frac{n_n}{n}\,\omega_p^2. \quad (7.23)$$

Thus the condition that μ^{-1} be finite as $k\to0$, which is equivalent to the condition that the current correlations have finite range, is also equivalent to the condition that the transverse conductivity sum rule be exhausted by the regular part of σ'^T.

When $k\neq0$, the two sides of (7.14) are equal, and the *regular part* of σ'^T satisfies the sum rule if and only if there is no magnetic susceptibility, i.e., when $\mu(k)=1$. This is the case in one special circumstance—a system with classical dynamics and with no intrinsic magnetic moments. In that case the current is purely orbital and, as first shown by van Leeuwen,[34] there is no orbital diamagnetism. To prove it in our formulation we note that the classical fluctuation dissipation theorem enables us to write

$$\int_{-\infty}^{\infty}\frac{d\omega}{\pi}\frac{\chi_{JJ}{}''^T(k\omega)}{\omega}=\beta\int d\mathbf{r}\,\exp(-i\mathbf{k}\cdot\mathbf{r})\,\langle j_k(\mathbf{r},0)j_l(\mathbf{0},0)\rangle(\delta_{kl}-k_kk_l/k^2)\tfrac{1}{2}.$$

The instantaneous velocity correlation is immediately evaluable classically and gives for the right-hand side ω_p^2. There is, of course, a paramagnetic contribution which alters this result even classically when we allow for intrinsic magnetization but use classical dynamics.

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[32] The general significance of such δ-function terms has been stressed in Ref. 21.

[33] P. C. Martin and J. Schwinger, Phys. Rev. **115**, 1355 (1959).

[34] J. H. Van Leeuwen, dissertation, Leiden, 1919 (unpublished); N. Bohr, dissertation, Copenhagen, 1911 (unpublished); as quoted in J. H. Van Vleck, *Classical Theory of Magnetic Susceptibilities* (Oxford University Press, London, 1932), Secs. 24–27.