

Memory function for the response of conduction electrons in metals

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The results of the quantum-mechanical treatment of the corrections to the dielectric function due to scattering are simulated by an extension of the Drude theory with a memory-function term. This phenomenological approach remains valid at frequencies of the order of the Fermi energy, where Drude's treatment fails. The optical conductivity and changes of the plasma frequency with scattering are discussed.

I. INTRODUCTION

The response of conduction electrons of metals to external fields is often treated with the Drude theory, where the scattering is described by a frequency-independent relaxation time. At high frequencies this method fails.¹ The corrections to the dielectric function due to scattering have been evaluated in the Born approximation.^{2,3} For low frequencies, principal diagrams of the perturbation series were summed.² This treatment leads to a frequency-dependent relaxation time in agreement with the observed optical conductivity.^{4,5} These results are obtained after a lengthy calculation, which hides the physical content. Here an extension of the Drude theory is introduced, which makes use of a memory function and leads to results similar to those of quantum-mechanical calculation.

The Drude theory is based on the classical equation of motion for particles of charge e and mass m in an electric field E with frequency ω ,

$$m\ddot{x} + m\xi_0\dot{x} = eEe^{-i\omega t}, \quad (1)$$

where ξ_0 is the reciprocal-relaxation time. This leads to a dielectric constant

$$\epsilon(\omega) = 1 - (\omega_p/\omega)^2(1 + i\xi_0/\omega)^{-1}, \quad (2)$$

where $\omega_p^2 = 4\pi Ne^2/m$, N being the density of the carriers. The corresponding result of the quantum mechanical treatment² is

$$\epsilon(\omega) = 1 - (\omega_p/\omega)^2[1 - \gamma(\omega) + i\xi(\omega)/\omega]^{-1}. \quad (3)$$

The functions $\gamma(\omega)$ and $\xi(\omega)$ are obtained from the pseudopotential V_q and the structure factor $S(q)$ of the scatterers through the expression^{2,6}

$$\begin{aligned} \gamma(\omega) - i\frac{\xi(\omega)}{\omega} &= \frac{k_F}{6\pi^4 N k_T^2 \omega^2} \int_0^\infty dq q^6 |V_q|^2 \\ &\times S(q) [\epsilon(q, 0) - \epsilon(q, \omega)], \end{aligned} \quad (4)$$

where $\epsilon(q, \omega)$ is Lindhard's complex dielectric function,⁷ $k_{TF} = (6\pi Ne^2/E_F)^{1/2}$ is the Thomas-Fermi screening parameter, and $E_F = k_F^2/2m$ is the Fermi energy. Units with $\hbar = 1$ are used.

In the limit $\omega \rightarrow 0$, Eq. (4) becomes

$$\begin{aligned} \gamma(0) &= \frac{m^2}{3\pi^4 N} \int_0^\infty dq q |V_q|^2 S(q) \\ &\times \left(\frac{1}{4} \ln \left| \frac{q + 2k_F}{q - 2k_F} \right| - \frac{qk_F}{(2k_F)^2 - q^2} \right), \end{aligned} \quad (5)$$

$$\xi(0) = \frac{m}{12\pi^3 N} \int_0^{2k_F} dq q^3 |V_q|^2 S(q), \quad (6)$$

and for $\omega \ll E_F$ Eq. (3) can be approximated by

$$\epsilon(\omega) = 1 - (\omega_p/\omega)^2 [1 - \gamma(0) + i\xi(0)/\omega]^{-1}. \quad (7)$$

This agrees with Drude's result, if we identify

$$\xi(0) = \xi_0 \quad (8)$$

and neglect $\gamma(0)$ compared to $\xi(0)/\omega$. The sign of $\gamma(0)$ depends on the function $|V_q|^2 S(q)$; it is negative for sodium.

For high frequencies, such that $\omega \geq q_0^2/2m$, $k_F q_0/m$ where q_0 is the range of $|V_q|^2$, the quantum result (4) shows that γ and ξ become frequency dependent. Therefore, Drude's formula cannot cover this frequency range with a constant ξ_0 .

II. MEMORY FUNCTION

The change of the relaxation frequency with the frequency of the external field can be incorporated into Drude's treatment with an additional term

$$m\ddot{x} + m\xi_0\dot{x} - \gamma_0 m \int dt' M(t-t')\dot{x}(t') = eEe^{-i\omega t}, \quad (9)$$

where $M(t)$ plays the role of a memory function, which is to be chosen in such a way that the quantum result is reproduced. Memory functions are often introduced into classical equations of motion to simulate frequency-dependent forces.⁸ An approach in which the quantum behavior of a Fer-

mi gas is described by a memory function has been proposed by Jindal *et al.*⁹ It is not transparent, however, how their memory function in the limit $q \rightarrow 0$ compares with that of Eqs. (13) or (14) and (15).

Equation (9) leads to a dielectric function

$$\epsilon(\omega) = 1 - (\omega_p/\omega)^2 [1 - \gamma_0 M(\omega) + i\xi_0/\omega]^{-1}. \quad (10)$$

This coincides with (3) with the choice

$$\text{Im}\gamma_0 M(\omega) = [\xi_0 - \xi(\omega)]/\omega, \quad (11)$$

$$\text{Re}\gamma_0 M(\omega) = \gamma(\omega). \quad (12)$$

This can be combined to

$$\gamma_0 M(\omega) = [\gamma(\omega) - i\xi(\omega)/\omega] + i\xi_0/\omega, \quad (13)$$

where the terms in the square brackets are given by (4). The memory function $\gamma_0 M(\omega)$ simply subtracts the Drude term in (10) and adds the quantum expression.

The detailed form of M depends on $|V_q|^2 S(q)$, which often may not be available. However, a general feature of the memory function is that it decays with a time τ_c . In view of this, in the absence of detailed information regarding $|V_q|^2 S(q)$, we may consider the approximate form

$$M(t) = \Theta(t) e^{-t/\tau_c}, \quad (14)$$

$$M(\omega) = (1 - i\omega\tau_c)^{-1}, \quad (15)$$

where $\Theta(t)$ is the step function and τ_c^{-1} is of the order of the Fermi energy. An exponential memory function gives a rather good, although not perfect, representation of the dynamics, even though it is in error at the origin ($t \ll \tau_c$). The form (14) for $M(t)$ used in the equation of motion (9) leads for short times to an additional relaxation term $\gamma_0 m \dot{x}/\tau_c$, while it becomes negligible for long times. Equations (11), (12), and (15) imply the frequency dependences

$$\xi(\omega) = \xi_0 - \gamma_0 \tau_c \omega^2 / (1 + \omega^2 \tau_c^2), \quad (16)$$

$$\gamma(\omega) = \gamma_0 / (1 + \omega^2 \tau_c^2). \quad (17)$$

Actually, for a screened Coulomb potential the expression (4) leads to $\xi(\omega) \sim \omega^{-1/2}$ for $\omega \rightarrow \infty$,

while for a constant $|V_q|^2 S(q)$ the asymptotic behavior $\xi(\omega) \sim \omega^{1/2}$ results. The constant asymptotic value $\xi(\infty) = \xi_0 - \gamma_0/\tau_c$ of (16) somehow lies between these extremes. The quantum expression for $\gamma(\omega)$ behaves in these cases as ω^{-2} and $\omega^{-1/2}$, respectively. The two functions $\gamma(\omega)$ and $\xi(\omega)/\omega$ are connected by the Kramers-Kronig relations.

The optical conductivity $\sigma(\omega) = (\omega/4\pi) \text{Im}\epsilon(\omega)$ with (3), (16), and (17) becomes, for $\omega \gg \xi_0$ and $\gamma_0 \ll 1$,

$$\sigma(\omega) = \frac{1}{4\pi} \left(\frac{\omega_p}{\omega}\right)^2 \left(\xi_0 - \frac{\omega^2 \tau_c \gamma_0}{1 + \omega^2 \tau_c^2}\right). \quad (18)$$

The last term accounts for the discrepancy with the Drude theory.

For liquid sodium this formula reproduces the measured values of $\sigma(\omega)$ (Refs. 1 and 4) with $\omega_p = 5.9$ eV, $\tau_c^{-1} = 3.12$ eV ($= E_F$) using the fitted parameters $\xi_0 = 0.44 \times 10^{14}$ sec⁻¹ = 0.029 eV and $\gamma_0 = -0.026$. The limited range of the measurement determines τ_c^{-1} only within the interval 3–6 eV.

This paper does not discuss interband transitions. The memory effect already occurs in an electron gas with scatterers and a homogeneous positive background. If there are interband effects in metals, they add on top of the memory effect.

The dielectric function (3) vanishes at the true plasma frequency: $\epsilon(\tilde{\omega}_p) = 0$. For $\xi(\omega_p)/\omega_p \ll 1$ and $\gamma(\omega_p) \ll 1$ this leads with (16) and (17) to

$$\frac{\tilde{\omega}_p - \omega_p}{\omega_p} = \frac{1}{2} \left[\frac{\gamma_0}{1 + \omega_p^2 \tau_c^2} - i \left(\frac{\xi_0}{\omega_p} - \frac{\gamma_0 \omega_p \tau_c}{1 + \omega_p^2 \tau_c^2} \right) \right]. \quad (19)$$

It is noteworthy that there is a shift to first order in γ_0 , while the Drude theory to this approximation gives no change in the real part of $\tilde{\omega}_p$. A shift of more than 1 eV in the plasma frequency as a function of temperature has indeed been observed in Ni,¹⁰ and has been accounted for by an explicit calculation of $\gamma(\omega_p)$ using Eq. (4).⁶

The addition of a memory function to the Drude theory thus extends this phenomenological method into the high-frequency range. It may often be useful, when the quantum-mechanical calculation (4) is not feasible because $|V_q|^2$ or $S(q)$ are unknown.

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