# SCATTERING OF SLOW ELECTRONS IN A POLAR MEDIUM

R. R. Dogonadze and A. A. Kornyshev

UDC 541.13

1. At the present time there is great interest in the development of methods of measurement of the complex dielectric permeability tensor $\varepsilon_{\alpha\beta}(\omega, \underline{q})$ [1] of a polar medium as a function of the independent variables frequency $\omega$ and wave vector $\mathbf{q}$. Determination of this function makes it possible to use a phenomenological method in the thermodynamics of dielectrics [2, 3], the theory of polarons [4] and excitons [5], the kinetics of charge transfer in a polar liquid [6, 7] (in particular, in the quantum theory of chemical and electrochemical reactions [8]), and in the theory of solutions of electrolytes [9] where the function $\varepsilon_{\alpha\beta}(\omega,q)$ which itself contains information on the multiparticle properties of a polar medium, is involved in the theory as an unknown parameter. Measurement of $\varepsilon_{\alpha\beta}(\omega, \underline{q})$ also gives valuable information on the structure of the polar liquid.

A natural possibility for measurement of the dielectric function in the surface $(\omega, q)$ is a study of the scattering of a test particle in the fluctuating electromagnetic field of the medium, which is caused by fluctuations in polarization and polarization current, with change in energy $(p'^2 - p^2)/2m = \hbar\omega$ and impulse $p' - p = \hbar\underline{q}$ of the particle (to the degree of accuracy indicated in [10]). In fact, the scattering section is expressed by the intensity of these fluctuations which is directly connected with $\varepsilon_{\alpha\beta}(\omega, \underline{q})$ [3, 10]. The test particle must simultaneously satisfy the following requirement.

a) It should only be scattered in the electromagnetic field of the medium. In the presence of other interactions it is necessary to have some means of detecting the electromagnetic scattering against the background scattering caused by other interactions.

b) It should react weakly with the electromagnetic field of the medium. If this is not the case then perturbation theory is inapplicable to calculation of the section, i.e., if it can be expressed by $\varepsilon_{\alpha\beta}(\omega, \underline{q})$ it will be an extremely complex function.

c) It should be sufficiently stable and controllable.

Generally, electrons react strongly with polarization [requirement b) is not satisfied] and can form bound states (polarons) with the medium.* In the following discussion we will deal with the specific case of electron scattering when the interaction with the field of the medium may be considered small.

2. It is known [11] that in the natural frequency spectrum of the dielectric there are transparency zones, i.e., ranges of frequency in which there is no absorption of quanta. Suppose an electron enters with an energy corresponding to frequency $\omega_n$ in the center of a sufficiently wide transparency zone (n). The degrees of freedom of the medium with frequencies $\gg\omega_n$ will be termed a "fast" subsystem of the medium with regard to the electron. The degrees of freedom with frequencies $\ll\omega_n$ separated from these by the transparency zone are termed "slow." The essential feature here is that such an "intermediate" electron is not converted into a polaron. In fact, the rapid subsystem instantaneously forms a polar "coat." However, this "coat" is inertia-free and follows the movement of the electron adiabatically. Such a light "coat"

*The neutrons discussed in [10] interact weakly with the field of a nonmagnetic medium but react strongly with the nuclei of the atoms of the medium. The electromagnetic scattering of the neutrons may be separated from the background of nuclear scattering by measurement of the angular variation of the scattering at angles $\lesssim 1'$. Positrons and muons [10] satisfy the requirements a) and b) but do not satisfy c).

Institute of Electrochemistry, Academy of Sciences of the USSR, Moscow. Translated from Élektrokhimiya, Vol. 9, No. 9, pp. 1321-1325, September, 1973. Original article submitted January 10, 1973.

© 1974 Consultants Bureau, a division of Plenum Publishing Corporation, 227 West 17th Street, New York, N. Y. 10011. No part of this publication may be reproduced, stored in a retrieval system, or transmitted, in any form or by any means, electronic, mechanical, photocopying, microfilming, recording or otherwise, without written permission of the publisher. A copy of this article is available from the publisher for $15.00.

has practically no effect on the electron (i.e., change in effective mass, law of dispersion, etc., may be neglected). The slow subsystem cannot form a "coat" and in relation to the electron it behaves as a fixed external field. The probability of an electron obtaining energy from the slow subsystem which considerably exceeds its own frequency and of being transferred to the absorption zone is small since this corresponds to multiphonon processes. If in an experiment the energies of an electron in the injector and detector correspond to the same transparency zone n and differ by the value $\hbar\omega$ ($\ll \hbar\omega_n$) we will consider that such a weakly inelastic scattering in caused by the production (destruction) of a polarization phonon [3] of the slow subsystem with an energy equal to $\hbar\omega$, i.e., by single-stage scattering in the field of the slow subsystem.*

Considering electrons with a wavelength greater than $10^{-8}$ cm (with an energy less than 4 eV), we will write the energy operator for their interaction with the medium in the form $\hat{V}(\underline{r}) = e\hat{\Phi}_S(\underline{r})$, where $\hat{\Phi}_S(\underline{r})$ is the fluctuating potential of the slow subsystem. According to [10], for the differential section of unit volume of the medium we have, using the Born approximation

$$\frac{d^2\Sigma}{d\omega\, d\Omega} = \frac{m^2e^2}{\hbar(2\pi\hbar)^3}\frac{p'}{p}\int_{-\infty}^{\infty} d\tau \exp\{i\omega\tau\} \sum_{\nu}\left\langle \nu \left| \exp\left\{\frac{F-\hat{H}}{T}\right\}\hat{\Phi}_s^{+}(q,0)\,\hat{\Phi}_s(q,\tau)\right| \nu\right\rangle, \tag{1}$$

$$\hat{\Phi}_s(q,t) = \exp\left\{\frac{i}{\hbar}\hat{H}t\right\}\left[\frac{1}{\sqrt{V}}\int dr \exp\{-iqr\}\hat{\Phi}_s(r)\right]\exp\left\{-\frac{i}{\hbar}\hat{H}t\right\}, \tag{2}$$

where $\hat{H}$, F, V, and $\nu$ are the Hamiltonian, free energy, volume and index of the internal state of the medium [12]. In a homogeneous isotropic medium $\hat{\Phi}_S(\underline{q}, t) = -\hat{P}_S^{\parallel}(\mathbf{q}, t)4\pi i/q$, where $P_S^{\parallel}(\mathbf{q}, t)$ is the linear (with respect to $\mathbf{q}$) component of the polarization vector of the slow subsystem. Using the expansion of this value with respect to the polarization phonons, the method of the effective Hamiltonian, and the rule of summation with respect to optical branches [3], from (1) we obtain

$$\frac{d^2\Sigma}{d\omega\, d\Omega} = \frac{mp'}{\pi^2\hbar r_B q^2 p}\left[\exp\left\{\frac{\hbar\omega}{T}\right\}-1\right]^{-1}\frac{\operatorname{Im}\varepsilon^{\parallel}(\omega,q)}{|\varepsilon^{\parallel}(\omega,q)|^2}\theta(\omega_n-\omega); \tag{3}$$

$r_B$ is the Boh radius.† Within the range of its applicability ($\hbar\omega \ll \hbar\omega_n$), Eq. (3) may be written in the form:

$$\frac{d^2\Sigma}{d\omega\, d\Omega} \simeq \frac{1+\dfrac{\hbar\omega}{mv_n^2}}{4\pi^2\sin^2\dfrac{\theta}{2}}\frac{e^2}{\hbar v_n}\frac{1}{v_n}\left[\exp\left\{\frac{\hbar\omega}{T}\right\}-1\right]^{-1}\operatorname{Im}\left\{-\varepsilon^{\parallel}\left[\omega, \frac{mv_n}{\hbar}\left(1+\frac{\hbar\omega}{2mv_n^2}\right)2\sin\frac{\theta}{2}\right]\right\}^{-1}, \tag{4}$$

where $v_n = p/m = \sqrt{2\hbar\omega_n/m}$. By experimental variation of the scattering angle $\theta$ and $\omega$ ($<\omega_n$), it is possible to determine $\operatorname{Im}\{\varepsilon^{\parallel}\}^{-1}$ over a wide range of $\omega$ and q with the aid of Eq. (4).

3. For a specific case let us discuss the scattering at an angle $\theta \lesssim 1$ of a beam of electrons falling perpendicularly on the plane surface of a sample of thickness L. The scattering probability density in unit time $d^3W/dtd\omega d\Omega = L \cdot dN/dt \cdot d^2\Sigma/d\omega d\Omega$, where dN/dt is the number of particles falling on the surface of the sample per unit time. Then the probability density of particles scattering in a volume angle $\Omega$ with change in energy $\hbar\omega$, $d^3W/dNd\omega d\Omega = L \cdot d^2\Sigma/d\omega d\Omega$. Let us consider scattering with change in energy in the range $\hbar\omega - \hbar\omega + \hbar\Delta\omega$. The smallness of the value

$$\frac{d^3W}{dN\, d\omega\, d\Omega}|\Delta\omega| \sim \frac{1}{\pi}\frac{|\omega|}{\omega}\frac{e^2}{\hbar v_n}\left[\exp\left\{\frac{\hbar\omega}{T}\right\}-1\right]^{-1}\frac{\operatorname{Im}\varepsilon^{\parallel}(\omega,\ldots)}{|\varepsilon^{\parallel}(\omega,\ldots)|^2}$$

*In principle, the recorded result may be obtained by another means. For example, having obtained energy from many phonons of the slow subsystem, the electron transfers to the neighboring absorption zone and then, thermalizing, returns to the original transparency zone. The electron may also obtain an energy $\hbar\omega$ from several phonons of the slow subsystem. An attempt to establish the necessary criterion for smallness of the effects of multiple scattering will be made in paragraph 3.

†The $\theta$-function automatically indicates that as a result of single-stage scattering the slow system cannot give any energy greater than $\hbar\omega_n$ to an electron.

where $\omega_* \equiv v_n/L$ is a necessary condition for the possibility of use of perturbation theory. Data on the study of dielectric relaxation [11] and absorption of electromagnetic waves [13] makes it possible to find the values $\varepsilon(\omega, q = 0) \equiv \varepsilon(\omega)$. For this reason, using the inequality [3]

$$\operatorname{Im} \varepsilon^{-1}(\omega, q) / |\varepsilon^{-1}(\omega, q)|^2 \leqslant \operatorname{Im} \varepsilon(\omega) / |\varepsilon(\omega)|^2;$$

we will write the required criterion in the form

$$\frac{1}{2\pi} \frac{L}{r_B} \frac{|\omega|}{\omega_n} \left[ \exp\left\{\frac{\hbar\omega}{T}\right\} - 1 \right]^{-1} \frac{\operatorname{Im} \varepsilon(\omega)}{|\varepsilon(\omega)|^2} \ll 1. \tag{5}$$

If $\hbar\omega_n > T$ then the slow subsystem involves both classical and quantum degrees of freedom of the medium (with particles less and greater than T respectively). Then (5) for scattering in classical polarization ($\hbar\omega \ll T$) has the form

$$\frac{1}{2\pi} \frac{T}{\hbar\omega_n} \frac{L}{r_B} \frac{\operatorname{Im} \varepsilon(\omega)}{|\varepsilon(\omega)|^2} \ll 1; \tag{6}$$

in the quantum case $\hbar\omega \gg T$

$$\frac{1}{2\pi} \frac{\omega}{\omega_n} \exp\left\{-\frac{\hbar\omega}{T}\right\} \frac{L}{r_B} \frac{\operatorname{Im} \varepsilon(\omega)}{|\varepsilon(\omega)|^2} \ll 1, \quad \omega > 0, \tag{7a}$$

scattering with absorption of a phonon of energy $\hbar\omega$,

$$\frac{1}{2\pi} \frac{\omega}{\omega_n} \frac{L}{r_B} \frac{\operatorname{Im} \varepsilon(\omega)}{|\varepsilon(\omega)|^2} \ll 1, \quad \omega < 0, \tag{7b}$$

and scattering with production of a phonon of energy $\hbar\omega$. From (5), (6), (7) it is possible to estimate the allowable thickness of the sample. For example, in water under normal conditions in experiments on scattering in the classical subsystem $L \ll 10^{-19}\omega_n$ cm, i.e., $L \ll 10^{-3}$ cm and in using a transparency zone with $\omega_n \sim 10^{16}$ sec$^{-1}$, $L \ll 10^{-5}$ cm when $\omega_n \sim 10^{14}$ sec$^{-1}$. For scattering in a quantum subsystem with energy gain $L \ll 10$ cm and with energy loss $L \ll 10^{-6}\omega_n/\omega$.

The greater the energy of the electron, i.e., the higher the frequency at the transparency zone used, the wider the range of applicability of perturbation theory (the greater the allowable thickness of the sample) and the zone of independent change in $\omega$ and q [10]. However, the greater the energy of the electrons in comparison to T, the more readily are they controlled. If precision recording of small changes in energy and impulse is practically feasible then high-energy zones of transparency for measurement of $\varepsilon$ at all frequencies should be used.

It should be emphasized that for a determination of $\varepsilon$ at quantum frequencies it is possible to conduct experiments on sufficiently thick samples (L = 1 cm), measuring the scattering of the electron with increase in its energy.

The authors express their thanks to Yu. A. Vdovin and M. I. Ryazanov for discussion of the results of the investigation.

## LITERATURE CITED

1. V. P. Silin and A. A. Rukhadze, The Electromagnetic Properties of Plasma and Plasma-Like Media [in Russian], Atomizdat (1961), Chap. 1.
2. A. A. Abrikosov, L. P. Gor'kov, and I. E. Dzyaloshinskii, Methods of Quantum Field Theory in Statistical Physics [in Russian], GIFML, Moscow (1962).
3. R. R. Dogonadze, A. A. Kornyshev, and A. M. Kuznetsov, Teor. i Matem. Fiz., 15, 58 (1973).
4. I. G. Lang and I. A. Firsov, Zh. Éksperim. i Teor. Fiz., 43, 1843 (1962); A. A. Ovchinnikov and M. Ya. Ovchinnikova, Teor. i Matem. Fiz., 7, 129 (1971); R. R. Dogonadze, Ber. Bunsenges. phys. Chem., 75, 628 (1971).
5. A. S. Davydov, The Theory of Molecular Excitons [in Russian], Nauka, Moscow (1968).
6. R. R. Dogonadze, A. M. Kuznetsov, and M. A. Vorotyncev, Phys. St. Sol. (b), 54, 125, 425 (1972).
7. R. R. Dogonadze and A. A. Kornyshev, Phys. St. Sol., (b) 53, 439 (1972); 55, 843 (1973).
8. A. M. Kuznetsov, Dissertation, Institute of Electrochemistry, Moscow (1971).
9. R. R. Dogonadze and A. A. Kornyshev, Dokl. Akad. Nauk SSSR, 207, 896 (1972).
10. R. R. Dogonadze and A. A. Kornyshev, Phys. St. Sol. (b), 49, 453 (1972).

11. H. Fröhlich, The Theory of Dielectrics, Oxford Univ. Press (1958).
12. L. Van Hove, N. M. Hugenholtz, and L. P. Howland, Quantum Theory of Many-Particle Systems, Benjamin (1961).
13. R. V. Pol', An Introduction to Optics, OGIZ-GOSTEKhIZDAT, Moscow (1947).