Dynamic Structure Factor: An Introduction*

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Z. Naturforsch. **48a**, 233–242 (1993); received December 10, 1991

The doubly differential cross-section for weak inelastic scattering of waves or particles by many-body systems is derived in Born approximation and expressed in terms of the dynamic structure factor according to van Hove. The application of this very general scheme to scattering of neutrons, x-rays and high-energy electrons is discussed briefly. The dynamic structure factor, which is the space and time Fourier transform of the density-density correlation function, is a property of the many-body system independent of the external probe and carries information on the excitation spectrum of the system.

The relation of the electronic structure factor to the density-density response function defined in linear-response theory is shown using the fluctuation-dissipation theorem. This is important for calculations, since the response function can be calculated approximately from the independent-particle response function in self-consistent field approximations, such as the random-phase approximation or the local-density approximation of the density functional theory. Since the density–density response function also determines the dielectric function, the dynamic structure can be expressed by the dielectric function.

Key words: Scattering theory; Dynamic structure factor; Inelastic photon scattering; Inelastic electron scattering; Inelastic neutron scattering; Dielectric function.

I. Introduction

Scattering of neutrons, high energy electrons, and photons by many-body systems are the most important methods for obtaining information on the spatial structure and on the dynamical structure, i.e., on the excitation spectrum, of the many-body system in question. Whereas the spatial structure is derived from elastic scattering, the excitations of the system are monitored by analyzing the inelastically scattered particles.

Neutrons "see" predominantly the nuclei, electrons and photons "see" predominantly the electrons in the system. Coupling to magnetic moments will not be considered in this article. More precisely, the information on the spatial structure is obtained by analyzing the scattered particles with respect to the scattering angle, i.e., with respect to change of the momentum. A measure of the scattering intensity is the differential cross-section

$$\mathrm{d}\sigma/\mathrm{d}\Omega\,.$$

The dynamic structure is obtained by analyzing the scattered particles with respect to scattering angle and kinetic energy. A measure of the inelastically scattered particles is the doubly differential cross-section

$$\mathrm{d}^2\sigma/\mathrm{d}\Omega\,\mathrm{d}E_1\,,$$

which is the subject of this article.

After a general derivation in the weak scattering limit, the doubly differential cross-section is worked out for the scattering of x-rays, fast electrons, and neutrons. Following van Hove, it is shown that the scattering intensity is essentially determined by the scattering function or dynamic structure factor, which is the space and time Fourier transform of the density-density correlation function, a property of the unperturbed system.

A calculation of the dynamic structure factor of the electronic structure of the system requires the knowledge of the many-body eigenstates of the unperturbed system, i.e., the solution of the corresponding many-body Schrödinger equation. This is seldom possible. In order to apply approximate methods based on the solution of an effective *single*-particle Schrödinger equation, contact is first made with the density–density response function, defined in linear-response theory, using the fluctuation-dissipation (FD) theorem. The density–density response function also provides the microscopic definition of the macroscopic dielectric function. In the self-consistent-field (SCF) approximation, the response function of seemingly independent particles is used to find an approximation to the true

* Presented at the Sagamore X Conference on Charge, Spin, and Momentum Densities, Konstanz, Fed. Rep. of Germany, September 1–7, 1991.

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response function by considering the response of these particles to an effective microscopic field (mean field) that is linked self-consistently to the induced density fluctuations. In order to illustrate the SCF approximation, we calculate the response function for the model of a homogeneous electron gas and discuss briefly its excitation spectrum. The response function of a crystalline solid is formally derived in the SCF approximation based on the solution of the corresponding band structure problem. Finally, we briefly discuss the response of highly localized electrons.

II. The Doubly Differential Cross-Section

The doubly differential cross-section is the relative intensity of the inelastically scattered particles, more precisely [1]

[number of particles scattered per sec into a solid angle $\mathrm{d}\Omega$ in the direction $\hat{\boldsymbol{l}} = \boldsymbol{r}/r$ with energies between E_1 and $E_1 + \mathrm{d}E_1$]/[number of incoming particles per sec and per cm^2 along the z-direction with momentum $\hbar \boldsymbol{k}_0$ and energy E_0] $= N\,\mathrm{d}^2\sigma$,

where N is the number of scatterers in the target, or

$$\mathrm{d}^2\sigma = \frac{1}{N} \frac{\boldsymbol{j}_1(\boldsymbol{r}, \boldsymbol{k}_1, E_1) \cdot \hat{\boldsymbol{l}} r^2 \,\mathrm{d}\Omega \,\mathrm{d}E_1}{(j_0)_z}. \tag{1}$$

Here $\boldsymbol{j}_1(\boldsymbol{r}, \boldsymbol{k}_1, E_1)$ is the current density of the scattered particles at $\boldsymbol{r}$ with momentum $\boldsymbol{k}_1$ and energy E_1. In the denominator of (1), $(j_0)_z$ is the current density of the monochromatic beam in the z-direction (see Figure 1).

In quantum-mechanical terms, the numerator of (1) is given by the number of particles that are scattered into a group of states in a phase-space volume of d^3k_1 around $\boldsymbol{k}_1$:

$$N_0 \sum_{n_0, n_1} P_{n_0} w(n_0, \boldsymbol{k}_0 \to n_1, \boldsymbol{k}_1) D(\boldsymbol{k}_1) \,\mathrm{d}^3 k_1. \tag{2}$$

Here $D(\boldsymbol{k}_1)$ is the density of plane-wave states in $\boldsymbol{k}$-space, which for periodic boundary conditions in volume V is

$$D(\boldsymbol{k}_1) = V/(2\pi)^3$$

and

$$D(\boldsymbol{k}_1)\,\mathrm{d}^3k_1 = \frac{V}{(2\pi)^3} k_1^2 \,\mathrm{d}\Omega \,\mathrm{d}k_1.$$

N_0 is the number of particles in state $|\boldsymbol{k}_0\rangle$. The scattering system is characterized by the quantum numbers n_0 before and n_1 after scattering.

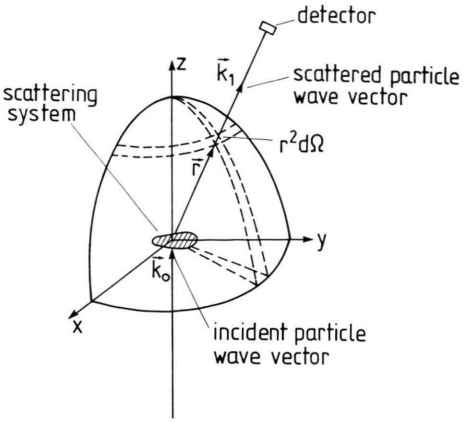

Fig. 1. Scattering geometry.

$w(n_0, \boldsymbol{k}_0 \to n_1, \boldsymbol{k}_1)$ is the transition rate for the particle to be scattered from the initial state $|\boldsymbol{k}_0\rangle$ to the final state $|\boldsymbol{k}_1\rangle$ while the system undergoes a transition from state $|n_0\rangle$ to state $|n_1\rangle$. P_{n_0} is the probability of finding the system in state $|n_0\rangle$ before scattering. At thermal equilibrium it is given by

$$P_{n_0} = e^{-\beta E_{n_0}}/Z,$$

where $H_0|n_0\rangle = E_{n_0}|n_0\rangle$, $Z = \sum_{n_0} e^{-\beta E_{n_0}}$ is the partition function and $\beta = 1/(k_B T)$. Before and after scattering particle and system do not interact, i.e. for $i = 0, 1$

$$|n_i, \boldsymbol{k}_i\rangle = |n_i\rangle\,|\boldsymbol{k}_i\rangle. \tag{3}$$

Assuming a weak interaction during scattering, the transition rate can be calculated in the Born approximation using (3),

$$w(n_0, \boldsymbol{k}_0 \to n_1, \boldsymbol{k}_1) \tag{4}$$

$$= \frac{2\pi}{\hbar} |\langle n_0, \boldsymbol{k}_0 | H_{\text{inter}} | n_1, \boldsymbol{k}_1 \rangle|^2 \,\delta(E_{n_0} + E_0 - E_{n_1} - E_1),$$

where H_{inter} is the interaction Hamiltonian with the system. Further evaluation of the doubly differential cross-section requires specification of the particle and its interaction with the system.

III. Inelastic X-Ray Scattering Spectroscopy (IXSS)

In X-ray scattering [2], the interaction Hamiltonian (in nonrelativistic approximation) consists of two terms ($-e$ is the charge of the electron):

$$H_{\text{inter}} = \frac{e}{mc} \sum_j \boldsymbol{A}(\boldsymbol{r}_j) \boldsymbol{p}_j + \frac{e^2}{2mc^2} \sum_j \boldsymbol{A}^2(\boldsymbol{r}_j). \tag{5}$$

The first term, which is linear in the vector potential $\boldsymbol{A}(\boldsymbol{r})$ that represents the photon field, describes in lowest order one-photon processes such as absorption and emission. Scattering is a two-photon process. The second order contribution of the first term is, for $\hbar\omega_0 \approx 10$ keV, smaller by $\hbar\omega_0/mc^2 \ll 1$ than the first-order contribution of the second term of (5), provided $\hbar\omega_0 \gg E_{n_1} - E_{n_0}$, i.e., in the nonresonant limit. Thus, if only the second term is important, the evaluation [3] of the transition probability yields

$$w(n_0, \boldsymbol{k}_0 \to n_1, \boldsymbol{k}_1) = \frac{2\pi}{\hbar}\left(\frac{2\pi c^2 \hbar}{V\sqrt{\omega_0 \omega_1}}\right)^2 r_0^2 (\boldsymbol{\varepsilon}_0 \cdot \boldsymbol{\varepsilon}_1)^2 |\langle n_1| \sum_j e^{i\boldsymbol{k}\cdot\boldsymbol{r}_j} |n_0\rangle|^2 \cdot \delta(E_{n_0} - E_{n_1} + \hbar\omega), \tag{6}$$

where $r_0 = e^2/(mc^2) = 2.8 \cdot 10^{-13}$ cm is the classical electron radius and $\boldsymbol{\varepsilon}_0$, $\boldsymbol{\varepsilon}_1$ are the polarization vectors of the incoming and outgoing photons, respectively. The momentum transfer $\boldsymbol{k}$ is defined by

$$\boldsymbol{k} = \boldsymbol{k}_0 - \boldsymbol{k}_1 \tag{7}$$

and the energy transfer $\hbar\omega$ by

$$\hbar\omega = E_0 - E_1 . \tag{8}$$

Since for photons $E_i = \hbar\omega_i = \hbar k_i c$ for $i = 0, 1$, $\mathrm{d}E_1 = \hbar c\, \mathrm{d}k_1$ and $(j)_z = (N_0/V) \cdot c$, we obtain finally

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\, \mathrm{d}E_1} = \left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_{\mathrm{Th}} \frac{k_1}{k_0} \frac{1}{N} \sum_{n_0, n_1} P_{n_0} |\langle n_1| \sum_j e^{i\boldsymbol{k}\boldsymbol{r}_j} |n_0\rangle|^2 \cdot \delta(E_{n_0} - E_{n_1} + \hbar\omega), \tag{9}$$

where

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_{\mathrm{Th}} = r_0^2 (\boldsymbol{\varepsilon}_0 \cdot \boldsymbol{\varepsilon}_1)^2 \tag{10}$$

is the Thomson cross-section for the scattering of a photon by an electron.

IV. Electron Energy Loss Spectroscopy (EELS)

In electron energy loss spectroscopy [2], as the inelastic scattering of high-energy electrons ($E_0 \approx 10$–100 keV) is called, the interaction Hamiltonian is given by the Coulomb interaction of the external electron at the position $\boldsymbol{r}$ and of the electrons in the system at the positions $\boldsymbol{r}_j$, i.e.,

$$H_{\mathrm{inter}} = e^2 \sum_j \frac{1}{|\boldsymbol{r} - \boldsymbol{r}_j|} . \tag{11}$$

Since for electrons $E_i = \dfrac{\hbar^2 k_i^2}{2m}$ for $i = 0, 1$, $\mathrm{d}E_1 = \dfrac{\hbar^2 k_1}{m}\mathrm{d}k_1$ and $(j_0)_z = \dfrac{N_0}{V} \cdot \dfrac{\hbar k_0}{m}$, we obtain

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\, \mathrm{d}E_1} = \left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_{\mathrm{R}} \frac{k_1}{k_0} \frac{1}{N} \sum_{n_0, n_1} P_{n_0} |\langle n_1| \sum_j e^{i\boldsymbol{k}\boldsymbol{r}_j} |n_0\rangle|^2 \cdot \delta(E_{n_0} - E_{n_1} + \hbar\omega), \tag{12}$$

where

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_{\mathrm{R}} = \frac{4m^2 e^4}{\hbar^4 k^4} \tag{13}$$

is the Rutherford cross-section for scattering of charged particles. The $1/k^4$ dependence is a consequence of the Coulomb interaction. Apart from the prefactor

$$\left(\frac{\mathrm{d}\sigma}{\mathrm{d}\Omega}\right)_{\mathrm{Th,R}} \cdot \frac{k_1}{k_0},$$

which accounts for the fundamental scattering process of the scattered particle by a single scatterer in the system, (9) and (12) are identical and dependent only on the properties of the scattering system. In other words, both EELS and IXSS yield the same information on the dynamical properties of the electronic system, i.e. on the excitation spectrum of the system, namely on transitions of the system from state $|n_0\rangle$ to state $|n_1\rangle$, provided

$$E_{n_1} - E_{n_0} = \hbar\omega ,$$

with the transition probability proportional to

$$|\langle n_1| \sum_j e^{i\boldsymbol{k}\boldsymbol{r}_j} |n_0\rangle|^2 .$$

The doubly differential cross-section yields an average over all possible excitations of the system with a fixed energy $\hbar\omega$ and a wavevector $\boldsymbol{k}$.

V. Inelastic Neutron Scattering

In neutron scattering [4], a neutron interacts with the nuclei at $\boldsymbol{r}_j$ in the system by a contact potential, the so-called Fermi pseudopotential,

$$H_{\mathrm{inter}} = \frac{2\pi\hbar}{m_n} \sum_j a_j \delta(\boldsymbol{r} - \boldsymbol{r}_j), \tag{14}$$

where m_n is the mass of the neutron and a_j is the scattering length for a neutron by a nucleus at $\boldsymbol{r}_j$,

which depends on the chemical and isotopical species.

For neutrons we have $E_i = \frac{\hbar^2 k_i^2}{2m_n}$ for $i = 0, 1$, $\mathrm{d}E_1 = \frac{\hbar^2 k_1}{m_n} \cdot \mathrm{d}k_1$, $(j_0)_z = \frac{N_0}{V} \cdot \frac{\hbar k_0}{m_n}$, and we obtain

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E_1} = \frac{k_1}{k_0}\frac{1}{N}\sum_{n_0,n_1} P_{n_0}|\langle n_1|\sum_j a_j e^{i\boldsymbol{k}\boldsymbol{r}_j}|n_0\rangle|^2 \cdot \delta(E_{n_0} - E_{n_1} + \hbar\omega). \tag{15}$$

VI. Correlation Function and Dynamic Structure Factor

(a) In EELS and in IXSS we observed that

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E_1} \sim \frac{1}{N}\sum_{n_0,n_1} P_{n_0}|\langle n_1|\sum_j e^{i\boldsymbol{k}\boldsymbol{r}_j}|n_0\rangle|^2 \cdot \delta(E_{n_0} - E_{n_1} + \hbar\omega) \equiv \frac{1}{\hbar}S(\boldsymbol{k},\omega), \tag{16}$$

where the scattering function or dynamic structure factor $S(\boldsymbol{k},\omega)$ depends on the properties of the unperturbed system only. Van Hove was the first to point out that $S(\boldsymbol{k},\omega)$ is the space and time Fourier transform of the density-density correlation function

$$\frac{1}{N}\langle \hat{n}(\boldsymbol{r},t)\,\hat{n}(\boldsymbol{r}',0)\rangle_T \tag{17}$$

where for an operator $\hat{A}$

$$\langle \hat{A}\rangle_T = \sum_{n_0} \frac{e^{-\beta E_{n_0}}}{Z}\langle n_0|\hat{A}|n_0\rangle.$$

The density-density correlation function is the probability of finding a particle at $\boldsymbol{r}$ at time t when there is one at $\boldsymbol{r}'$ at time $t = 0$. The following four steps provide the proof of van Hove's assertion [2]:

(i) The density operator $\hat{n}(\boldsymbol{r})$ is defined by

$$\hat{n}(\boldsymbol{r}) = \sum_j \delta(\boldsymbol{r} - \boldsymbol{r}_j) = \frac{1}{V_s}\sum_{\boldsymbol{k}} \hat{n}_{\boldsymbol{k}} e^{i\boldsymbol{k}\boldsymbol{r}}, \tag{18}$$

where the Fourier transform

$$\hat{n}_{\boldsymbol{k}} = \int d^3r\, \hat{n}(\boldsymbol{r}) e^{-i\boldsymbol{k}\boldsymbol{r}} = \sum_j e^{-i\boldsymbol{k}\boldsymbol{r}_j}. \tag{19}$$

(ii)

$$\delta(E_{n_0} - E_{n_1} + \hbar\omega) = \frac{1}{2\pi}\int \frac{\mathrm{d}t}{\hbar}\exp\{i(E_{n_0} - E_{n_1} + \hbar\omega)t/\hbar\}.$$

(iii)

$$\langle n_0|\hat{n}_{\boldsymbol{k}}|n_1\rangle \exp\{i(E_{n_0} - E_{n_1})t/\hbar\} = \langle n_0|e^{iE_{n_0}t/\hbar}\hat{n}_{\boldsymbol{k}}e^{-iE_{n_1}t/\hbar}|n_1\rangle = \langle n_0|e^{iH_0t/\hbar}\hat{n}_{\boldsymbol{k}}e^{-iH_0t/\hbar}|n_1\rangle = \langle n_0|\hat{n}_{\boldsymbol{k}}(t)|n_1\rangle,$$

where

$$\hat{n}_{\boldsymbol{k}}(t) = e^{iH_0t/\hbar}\hat{n}_{\boldsymbol{k}}\,\mathrm{e}^{-iH_0t/\hbar} \tag{20}$$

is the Heisenberg representation of the $\hat{n}_{\boldsymbol{k}}$.

(iv) Completeness relation:

$$\sum_{n_1}|n_1\rangle\langle n_1| = 1. \tag{21}$$

Using (i)–(iv) we find

$$\frac{1}{\hbar}S(\boldsymbol{k},\omega) = \frac{1}{N}\sum_{n_0,n_1} P_{n_0}|\langle n_1|\sum_j e^{i\boldsymbol{k}\boldsymbol{r}_j}|n_0\rangle|^2 \cdot \delta(E_{n_0} - E_{n_1} + \hbar\omega)$$
$$= \frac{1}{2\pi\hbar N}\int \mathrm{d}t\, e^{i\omega t}\sum_{n_0} P_{n_0}\langle n_0|\hat{n}_{\boldsymbol{k}}(t)\hat{n}_{-\boldsymbol{k}}(0)|n_0\rangle$$
$$= \frac{1}{2\pi\hbar N}\int \mathrm{d}t\, e^{i\omega t}\langle \hat{n}_{\boldsymbol{k}}(t)\hat{n}_{-\boldsymbol{k}}(0)\rangle_T \tag{22}$$
$$= \frac{1}{2\pi\hbar}\int \mathrm{d}t\, e^{i\omega t}\int \mathrm{d}^3r\int \mathrm{d}^3r'\, e^{-i\boldsymbol{k}(\boldsymbol{r}-\boldsymbol{r}')}\frac{1}{N}\langle \hat{n}(\boldsymbol{r},t)\hat{n}(\boldsymbol{r}',0)\rangle_T,$$

i.e. photons (X-rays) or high-energy electrons are scattered by the electron density fluctuations in the system.

(b) Using (i)–(iv), the neutron scattering cross-section becomes

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E_1} = \frac{k_1}{k_0}\frac{1}{2\pi\hbar N}\int \mathrm{d}t\, e^{i\omega t}\left\langle \sum_{j,j'} a_{j'}e^{-i\boldsymbol{k}\boldsymbol{r}_{j'}}a_j e^{i\boldsymbol{k}\boldsymbol{r}_j}\right\rangle_T. \tag{23}$$

For simplicity we consider systems comprising one chemical species only, but with different isotopes (assumed to be randomly distributed). We introduce an average scattering length

$$\overline{a_j} = \bar{a}$$

and the local deviation from it,

$$\delta a_j = a_j - \bar{a}.$$

Then $\overline{\delta a_j} = 0$ and

$$\overline{\delta a_j\,\delta a_{j'}} = \delta_{j,j'}\overline{(\delta a_j)^2} = \delta_{j,j'}\overline{(a_j - \bar{a})^2} = \overline{a^2} - \bar{a}^2$$

with $\overline{a^2} = \overline{a_j^2}$. In this way we can split the scattering cross-section into a coherent and an incoherent part,

$$\frac{\mathrm{d}^2\sigma}{\mathrm{d}\Omega\,\mathrm{d}E_1} = \frac{k_1}{k_0}\frac{1}{4\pi\hbar}[\sigma_{\mathrm{coh}} S_{\mathrm{coh}}(\boldsymbol{k},\omega) + \sigma_{\mathrm{inc}} S_{\mathrm{inc}}(\boldsymbol{k},\omega)], \tag{24}$$

where

$$S_{\mathrm{coh}}(\boldsymbol{k},\omega) = \frac{1}{2\pi}\int \mathrm{d}t\, e^{i\omega t}\frac{1}{N}\langle \hat{n}_{\boldsymbol{k}}(t)\,\hat{n}_{-\boldsymbol{k}}(0)\rangle_T \tag{25}$$

is the coherent scattering function, which includes interference effects. $\sigma_{\mathrm{coh}} = 4\pi\bar{a}^2$ is the coherent neutron scattering cross-section.

$$S_{\mathrm{inc}}(\boldsymbol{k},\omega) = \frac{1}{2\pi}\int \mathrm{d}t\, e^{i\omega t}\frac{1}{N}\sum_j \langle e^{-i\boldsymbol{k}\boldsymbol{r}_j(t)}\, e^{i\boldsymbol{k}\boldsymbol{r}_j(0)}\rangle_T \tag{26}$$

is the Fourier transform of a self-correlation function that accounts for the incoherent scattering processes, i.e., without interference effects. $\sigma_{\mathrm{inc}} = 4\pi(\overline{a^2} - \bar{a}^2)$ is the incoherent neutron scattering cross-section. σ_{coh} and σ_{inc} are typically of the order of 10^{-24} cm^2. Again $S_{\mathrm{coh}}(\boldsymbol{k},\omega)$ and $S_{\mathrm{inc}}(\boldsymbol{k},\omega)$ are functions describing the dynamic properties of the unperturbed system.

The calculation of the dynamical structure factors is a very difficult problem. The required many-body eigenstates of the system cannot be calculated in general, and we must depend on approximations. For this purpose another important function that describes the dynamical properties of the system is introduced. It is the density-density response function, which is related to the dynamic structure factor and for which, for example, the self-consistent-field approximation is such an approximate scheme.

VII. Response Function

Consider a *weak* external potential $\varphi_{\mathrm{ext}}(\boldsymbol{r},t)$ acting on the electrons in the system. Then

$$\begin{aligned} H_{\mathrm{inter}} &= (-e)\sum_j \varphi_{\mathrm{ext}}(\boldsymbol{r}_j,t) \\ &= \int \mathrm{d}^3r \sum_j \delta(\boldsymbol{r}-\boldsymbol{r}_j)(-e)\,\varphi_{\mathrm{ext}}(\boldsymbol{r},t) \\ &= \int \mathrm{d}^3r\, \hat{n}(\boldsymbol{r})(-e)\,\varphi_{\mathrm{ext}}(\boldsymbol{r},t). \end{aligned} \tag{27}$$

In linear-response theory [2], the density response of the system, e.g. to the external potential, can be calculated by first-order perturbation theory. One finds

$$\delta n(\boldsymbol{r},t) = \int \mathrm{d}^3r' \int \mathrm{d}t\, \chi(\boldsymbol{r},\boldsymbol{r}';t-t')(-e)\,\varphi_{\mathrm{ext}}(\boldsymbol{r}',t'), \tag{28}$$

where

$$\chi(\boldsymbol{r},\boldsymbol{r}';t-t') = -i\,\Theta(t-t')\langle[\hat{n}(\boldsymbol{r},t),\hat{n}(\boldsymbol{r}',t')]\rangle_T/\hbar \tag{29}$$

is the density-density response function. The Θ-function ($\Theta(\tau) = 0$ for $\tau < 0$; $\Theta(\tau) = 1$ for $\tau \geqq 0$) guarantees *causality*, i.e., contributions to the induced density fluctuations $\delta n(\boldsymbol{r},t)$ at time t can only arise from perturbations, i.e., from $\varphi_{\mathrm{ext}}(\boldsymbol{r},t)$ for $t' \leqq t$. The square bracket in (29) denotes a commutator, i.e., $[\hat{A},\hat{B}] = \hat{A}\hat{B} - \hat{B}\hat{A}$. An important consequence of causality are the Kramers–Kronig relations [5, 6]

$$\mathrm{Re}\,\chi(\boldsymbol{r},\boldsymbol{r}';\omega) = \frac{1}{\pi}P\int \mathrm{d}\omega' \frac{\mathrm{Im}\,\chi(\boldsymbol{r},\boldsymbol{r}';\omega')}{\omega'-\omega}, \tag{30a}$$

$$\mathrm{Im}\,\chi(\boldsymbol{r},\boldsymbol{r}';\omega) = \frac{-1}{\pi}P\int \mathrm{d}\omega' \frac{\mathrm{Re}\,\chi(\boldsymbol{r},\boldsymbol{r}';\omega')}{\omega'-\omega}. \tag{30b}$$

Here P denotes a principle-value integral. Using the Kramers–Kronig relations, sum rules can be obtained from

$$\langle[\hat{n}(\boldsymbol{r},t),\hat{n}(\boldsymbol{r}',t')]\rangle_T = -\int \frac{\mathrm{d}\omega}{\pi}\mathrm{Im}\,\chi(\boldsymbol{r},\boldsymbol{r}';\omega)\,e^{-i\omega(t-t')} \tag{31}$$

(i) by differentiating (31) with respect to t,
(ii) by using Heisenberg's equation of motion

$$i\hbar\frac{\mathrm{d}}{\mathrm{d}t}\hat{n}(\boldsymbol{r},t) = [\hat{n}(\boldsymbol{r},t),H_0], \tag{32}$$

(iii) by taking the limit $t = t'$

$$\begin{aligned} &\langle[(i\hbar\,\mathrm{d}/\mathrm{d}t)^n\,\hat{n}(\boldsymbol{r},t),\hat{n}(\boldsymbol{r}',t')]\rangle|_{t=t'} \\ &\quad = \langle[[\ldots[\hat{n}(\boldsymbol{r},t),H_0],H_0]\ldots],\hat{n}(\boldsymbol{r}',t)]\rangle_T \\ &\quad = -\hbar^n\int\frac{\mathrm{d}\omega}{\pi}\omega^n\,\mathrm{Im}\,\chi(\boldsymbol{r},\boldsymbol{r}';\omega), \end{aligned} \tag{33}$$

provided the equal time commutators in (33) can be evaluated. A well-known example is the f-sum rule, which is the first frequency moment ($n = 1$) of $\mathrm{Im}\,\chi(\boldsymbol{r},\boldsymbol{r}';\omega)$.

VIII. Macroscopic Dielectric Function

In order to solve Maxwell's equation in the presence of matter one needs a constitutive relation between the dielectric displacement $\boldsymbol{D}$ and the electric field strength $\boldsymbol{E}$. If we deal with longitudinal fields, we

can represent them by scalar potentials

$$\boldsymbol{D} = -\nabla\varphi_{\text{ext}},$$
$$\boldsymbol{E} = -\nabla\varphi_{\text{tot}}.$$

The constitutive relation is

$$\boldsymbol{D} = \varepsilon_{\text{M}}\boldsymbol{E},$$

which is valid for the space and time Fourier transform. ε_{M} is the macroscopic dielectric function. In terms of the scalar potentials, the constitutive relation can be written [6]

$$\frac{1}{\varepsilon_{\text{M}}(\boldsymbol{k},\omega)} = \frac{\varphi_{\text{tot}}(\boldsymbol{k},\omega)}{\varphi_{\text{ext}}(\boldsymbol{k},\omega)}. \tag{34}$$

For the macroscopic potentials we have

$$\varphi_{\text{tot}}(\boldsymbol{k},\omega) = \varphi_{\text{ext}}(\boldsymbol{k},\omega) + \varphi_{\text{ind}}(\boldsymbol{k},\omega). \tag{35}$$

The induced potential $\varphi_{\text{ind}}(\boldsymbol{k},\omega)$ is related to the induced density fluctuations $n(\boldsymbol{k},\omega)$ by Poisson's equation

$$\varphi_{\text{ind}}(\boldsymbol{k},\omega) = -\frac{4\pi e}{k^2} n(\boldsymbol{k},\omega). \tag{36}$$

From linear-response theory we have

$$n(\boldsymbol{k}',\omega) = \chi(\boldsymbol{k}',\boldsymbol{k};\omega)(-e)\varphi_{\text{ext}}(\boldsymbol{k},\omega),$$

where using (29) (and with V_S as the quantization volume of the system), the Fourier transform of the density-density response function is defined by

$$\chi(\boldsymbol{k}',\boldsymbol{k};\omega) = \frac{-i}{\hbar V_S}\int \mathrm{d}^3r\, e^{-i\boldsymbol{k}'\boldsymbol{r}}\int \mathrm{d}^3r' \cdot e^{i\boldsymbol{k}\boldsymbol{r}'}\int_0^\infty \mathrm{d}\tau\, e^{i(\omega+i\delta)\tau}\langle[\hat{n}(\boldsymbol{r},\tau),\hat{n}(\boldsymbol{r}',0)]\rangle_T \tag{37}$$

and

$$n(\boldsymbol{k}',\omega) = \frac{1}{V_S}\int \mathrm{d}^3r\,\delta n(\boldsymbol{r},\omega)\, e^{-i\boldsymbol{k}'\boldsymbol{r}}.$$

This provides us with a *microscopic* definition of the *macroscopic* dielectric function $\varepsilon_{\text{M}}(\boldsymbol{k},\omega)$ in terms of the density-density response function

$$\frac{1}{\varepsilon_{\text{M}}(\boldsymbol{k},\omega)} = 1 + \frac{4\pi e^2}{k^2}\chi(\boldsymbol{k},\boldsymbol{k},\omega). \tag{38}$$

IX. The Fluctuation-Dissipation Theorem

Obviously there is a formal similarity between the dynamic structure factor $S(\boldsymbol{k},\omega)$ and the density-density response function $\chi(\boldsymbol{k},\boldsymbol{k};\omega)$. The link is provided by the fluctuation-dissipation (FD) theorem [2]. With $n = N/V_S$ it reads

$$S(\boldsymbol{k},\omega) = \frac{1}{\pi n}\frac{\hbar}{e^{-\beta\hbar\omega}-1}\operatorname{Im}\chi(\boldsymbol{k},\boldsymbol{k};\omega) \tag{39}$$

or, using the definition of the dielectric function (38),

$$S(\boldsymbol{k},\omega) = \frac{\hbar k^2}{4\pi^2 e^2 n}\frac{1}{1-e^{-\beta\hbar\omega}}\operatorname{Im}\left[\frac{-1}{\varepsilon_{\text{M}}(\boldsymbol{k},\omega)}\right]. \tag{39'}$$

It is important to note from (39') that the imaginary part of the negative *inverse* of the dielectric function is determined by the excitation spectrum of the system. $\operatorname{Im}[-1/\varepsilon_{\text{M}}(\boldsymbol{k},\omega)]$ is known as the energy-loss function. The name fluctuation-dissipation theorem derives from the fact that $S(\boldsymbol{k},\omega)$ determines the scattering intensity of particles by the density fluctuations in the system. One can show, on the other hand, that $\operatorname{Im}\chi(\boldsymbol{k},\boldsymbol{k};\omega)$ determines the amount of energy absorbed by the system when perturbed by an external potential.

The fluctuation-dissipation theorem is of great practical importance for the approximate evaluation of the dynamic structure factor. The true many-body eigenstates $E_{n_0}, |n_0\rangle$ cannot be calculated in most cases except for a fictitious system of non-interacting particles. However, we cannot expect that $S(\boldsymbol{k},\omega)$ evaluated for non-interacting particles is a reasonable approximation to the true dynamic structure factor, and the same is true for the density-density response function.

The response function $\chi^0(\boldsymbol{k},\boldsymbol{k};\omega)$ of seemingly independent particles (electrons), however, can be used to find approximations to the true $\chi(\boldsymbol{k},\boldsymbol{k};\omega)$ within the self-consistent-field (SCF) approximation such as the random-phase approximation (RPA) or the local density approximation (LDA) in density functional theory [7]. Thus

$$\chi^0 \xrightarrow{\text{SCF}} \chi \xrightarrow{\text{FD}} S. \tag{40}$$

X. Self-Consistent-Field Approximation for Jellium

In order to illustrate what is meant by the symbolic relation (40) we calculate SCF approximations to χ for the model system of a homogeneous electron gas (jellium). Because of translational invariance,

$$\chi(\boldsymbol{r},\boldsymbol{r}';\omega) = \chi(|\boldsymbol{r}-\boldsymbol{r}'|;\omega)$$

and

$$n(\boldsymbol{k},\omega) = \chi(\boldsymbol{k},\omega)(-e)\,\varphi_{\text{ext}}(\boldsymbol{k},\omega). \tag{41}$$

For jellium, χ depends on the modulus of $|\boldsymbol{k}| = k$ only. In the SCF approximation for $\chi(k,\omega)$ we consider the response of seemingly independent particles to an effective microscopic field,

$$n(\boldsymbol{k},\omega) = \chi^0(k,\omega)(-e)\,\varphi_{\text{tot}}(\boldsymbol{k},\omega) \tag{42}$$

with

$$\chi^0(k,\omega) = \frac{2}{V_S}\sum_{\boldsymbol{q}} \frac{f(E_{\boldsymbol{q}}) - f(E_{\boldsymbol{q}+\boldsymbol{k}})}{\hbar\omega + i\delta + E_{\boldsymbol{q}} - E_{\boldsymbol{q}+\boldsymbol{k}}}. \tag{43}$$

$E_{\boldsymbol{q}} = \hbar^2 q^2/2m$ are single-particle energies of the plane-wave states $|\boldsymbol{q}\rangle$, and $f(E_{\boldsymbol{q}})$ is the Fermi function. Self-consistency means that the *microscopic* total potential is related to the external potential and to the induced density in a similar way as in classical electrodynamics,

$$\varphi_{\text{tot}}(\boldsymbol{k},\omega) = \varphi_{\text{ext}}(\boldsymbol{k},\omega) + \varphi_{\text{ind}}(\boldsymbol{k},\omega). \tag{44}$$

In the LDA, e.g., we obtain

$$\begin{aligned} -e\,\varphi_{\text{ind}}(\boldsymbol{k},\omega) &= \frac{4\pi e^2}{k^2} n(\boldsymbol{k},\omega) + \frac{\mathrm{d}^2}{\mathrm{d}n^2}[n\,\varepsilon_{\text{xc}}(n)]\, n(\boldsymbol{k},\omega) \\ &= (v_k + v_{\text{xc}})\, n(\boldsymbol{k},\omega), \end{aligned} \tag{45}$$

where $v_k = 4\pi e^2/k^2$ and $v_{\text{xc}} = \dfrac{\mathrm{d}^2}{\mathrm{d}n^2}[n\,\varepsilon_{\text{xc}}(n)]$. $\varepsilon_{\text{xc}}(n)$ is the exchange-correlation energy per particle in jellium. If we neglect v_{xc}, which is a true quantum mechanical effect, we obtain the RPA. From (42), (44) and (45) we find

$$n(\boldsymbol{k},\omega) = \frac{\chi^0(k,\omega)}{1 - (v_k + v_{\text{xc}})\chi^0(k,\omega)}(-e)\,\varphi_{\text{ext}}(\boldsymbol{k},\omega). \tag{46}$$

Comparing (41) and (46) we observe

$$\chi_{\text{LDA}}(k,\omega) = \frac{\chi^0(k,\omega)}{1 - (v_k + v_{\text{xc}})\chi^0(k,\omega)} \tag{47}$$

or, neglecting xc-effects,

$$\chi_{\text{RPA}}(k,\omega) = \frac{\chi^0(k,\omega)}{1 - v_k\chi^0(k,\omega)}. \tag{48}$$

Using (38) we obtain for the dielectric function

$$\varepsilon_{\text{M}}^{\text{LDA}}(k,\omega) = 1 - \frac{v_k\chi^0(k,\omega)}{1 - v_{\text{xc}}\chi^0(k,\omega)} \tag{49}$$

or, introducing the so-called local field factor $G^0(k)$ of the homogeneous electron gas by

$$v_{\text{xc}} = -v_k G^0(k), \tag{50}$$

$$\varepsilon_{\text{M}}^{\text{LDA}}(k,\omega) = 1 - \frac{v_k\chi^0(k,\omega)}{1 + v_k G^0(k)\chi^0(k,\omega)}. \tag{49'}$$

Since in the LDA v_{xc} is a negative constant, $G^0(k) \sim k^2$. This has the proper behaviour for small k, but it diverges for large k although it should become a constant. Approximations that satisfy both the small- and the large-k requirements for $G^0(k)$ qualitatively were proposed by Hubbard [8], Singwi et al. [9], Utsumi and Ichimaru [10] and by many others [11]. These are nonlocal density approximations. In principle, $G^0(k)$ should also depend on frequency [12].

XI. Excitation Spectrum of the Homogeneous Electron Gas in RPA

It is well-known that the RPA is valid only in the high-density limit ($r_s \ll 1$, where r_s is defined by $4\pi(r_s a_0)^3/3 = 1/n$). Normal metals range between $r_s = 2$ and 6. Nevertheless, it is instructive to discuss the excitation spectrum of the jellium in the RPA [6],

$$\varepsilon_{\text{M}}^{\text{RPA}}(k,\omega) = 1 - v_k\chi^0(k,\omega) \equiv \varepsilon_{\text{L}}(k,\omega), \tag{51}$$

where $\varepsilon_{\text{L}}(k,\omega)$ is the well-known Lindhard dielectric function, which is easily evluated for $T = 0$ from (43). Its explicit form can be found in many textbooks.

According to (39') the excitation spectrum is determined by the energy-loss function, which for jellium in the RPA is

$$\text{Im}\,[-1/\varepsilon_{\text{L}}(k,\omega)].$$

As illustrated in Fig. 2 it consists of a broad continuum of (screened) particle–hole excitations and, for small k, of a discrete collective excitation, the plasmon. This exhibits a quadratic dispersion for $k \to 0$ and ceases to exist for $k > k_c$, the so-called plasmon cutoff wavevector, which is of the order of the Fermi wavevector k_{F}. In the long-wavelength limit the plasma frequency ω_{p} is determined by

$$\omega_{\text{p}}^2 = 4\pi n e^2/m \tag{52}$$

and is typically of the order of the Fermi energy E_{F}. It was pointed out before (see (9) and (12)) that EELS and IXSS yield the same information on the excitation spectrum. There is an important practical difference,

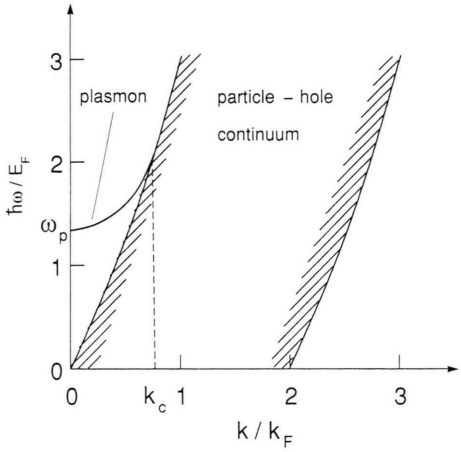

Fig. 2. Excitation spectrum of the homogeneous electron gas in the RPA for $r_s = 2.07$ (average valence electron density of aluminium).

however, because $(d\sigma/d\Omega)_{Th}$ is independent of k, whereas $(d\sigma/d\Omega)_R \sim 1/k^4$. Consequently

$$\left.\frac{d^2\sigma}{d\Omega\, dE_1}\right|_{EELS} \sim \frac{1}{k^2}\,\mathrm{Im}\left[\frac{-1}{\varepsilon_M(k,\omega)}\right]$$

favours small k, i.e., small-angle scattering, whereas

$$\left.\frac{d^2\sigma}{d\Omega\, dE_1}\right|_{IXSS} \sim k^2\,\mathrm{Im}\left[\frac{-1}{\varepsilon_M(k,\omega)}\right]$$

favours large k, i.e., large-angle scattering. EELS has been applied for many years for detailed studies of plasmons [13, 14]. Improvement in the experimental technique and the availability of synchrotron radiation sources [15] has led recently to the discovery of unexpected structures in the large-k regime of the particle–hole continuum that are particularly interesting in the context of xc-effects. This provided the motivation for very elaborate theoretical investigations of xc-effects on the dielectric function of the homogeneous electron gas [16]. Experimental investigations centred on simple metals and semiconductors, regarded as nature's closest realization of the jellium model.

One must keep in mind, however, that the effective crystal potential (pseudopotential), although rather small in simple metals and semiconductors, changes the simple picture derived from jellium considerably. A coupling of the two types of excitations via umklapp processes leads to a decay of plasmons by interband transitions and to plasmon bands, for example [17, 18]. The Bloch nature of the electronic states can lead to new collective modes (zone-boundary collective states [19–23]). The understanding of these properties is based on the SCF approximation for the density-density response function of crystalline solids [24, 25], which is outlined in the last section.

XII. Density Response in Crystalline Solids

Periodicity requires that

$$\chi(\boldsymbol{r}+\boldsymbol{R},\boldsymbol{r}'+\boldsymbol{R};\omega) = \chi(\boldsymbol{r},\boldsymbol{r}';\omega) \tag{53}$$

for any lattice vector $\boldsymbol{R}$. Expansion in a double Fourier series yields

$$\chi(\boldsymbol{r},\boldsymbol{r}';\omega) = \sum_{\boldsymbol{k},\boldsymbol{k}'} \chi(\boldsymbol{k},\boldsymbol{k}';\omega)\, e^{i\boldsymbol{k}\boldsymbol{r}}\, e^{-i\boldsymbol{k}'\boldsymbol{r}'}. \tag{54}$$

Because of (53) we must satisfy

$$e^{i(\boldsymbol{k}-\boldsymbol{k}')\boldsymbol{R}} = 1,$$

which requires

$$\boldsymbol{k} - \boldsymbol{k}' = \boldsymbol{G},$$

where $\boldsymbol{G}$ is any reciprocal-lattice vector. If we introduce the Brillouin zone (BZ), then

$$\boldsymbol{k} \to \boldsymbol{k} + \boldsymbol{G},$$

where $\boldsymbol{k}$ is now restricted to the first BZ. Then (54) becomes

$$\chi(\boldsymbol{r},\boldsymbol{r}';\omega) = \sum_{\boldsymbol{k},\boldsymbol{G},\boldsymbol{G}'} \chi(\boldsymbol{k}+\boldsymbol{G},\boldsymbol{k}+\boldsymbol{G}';\omega) \cdot e^{i(\boldsymbol{k}+\boldsymbol{G})\boldsymbol{r}}\, e^{-i(\boldsymbol{k}+\boldsymbol{G}')\boldsymbol{r}'}. \tag{54'}$$

This means, for example, that

$$n(\boldsymbol{k}+\boldsymbol{G},\omega) = \chi(\boldsymbol{k}+\boldsymbol{G},\boldsymbol{k};\omega)(-e)\,\varphi_{ext}(\boldsymbol{k},\omega) \tag{55}$$

is valid for any reciprocal-lattice vector $\boldsymbol{G}$. An external potential $\varphi_{ext}(\boldsymbol{k},\omega)\, e^{i\boldsymbol{k}\boldsymbol{r}}$ will not only excite density fluctuations of the same periodicity as the external potential has, but a whole spectrum of short-wavelength fluctuations and vice versa. According to (54') the response in a crystalline solid is represented in k-space by a response matrix χ spanned by reciprocal-lattice vectors $\boldsymbol{G}, \boldsymbol{G}'$.

Again we do not know the many-body eigenstates of the crystalline solid. Most calculations rely on the SCF approximation based on the solution of the corresponding band structure problem, i.e., one calculates the response matrix χ from the single-particle

Bloch states and band energies

$$\chi^0(\boldsymbol{k}+\boldsymbol{G},\boldsymbol{k}+\boldsymbol{G}';\omega)=\frac{2}{V_s}\sum_{\boldsymbol{q},l,l'}\langle l\boldsymbol{q}|e^{-i(\boldsymbol{k}+\boldsymbol{G})\boldsymbol{r}}|l',\boldsymbol{q}+\boldsymbol{k}\rangle \cdot\frac{f(E_{l\boldsymbol{q}})-f(E_{l',\boldsymbol{q}+\boldsymbol{k}})}{\hbar\omega+i\delta+E_{l\boldsymbol{q}}-E_{l',\boldsymbol{q}+\boldsymbol{k}}}\langle l',\boldsymbol{q}+\boldsymbol{k}|e^{i(\boldsymbol{k}+\boldsymbol{G}')\boldsymbol{r}}|l\boldsymbol{q}\rangle, \quad (56)$$

where the crystal momentum $\boldsymbol{q}$ is restricted to the first BZ and the band indices l (or l', respectively) characterize the Bloch states $|l\boldsymbol{q}\rangle$ and band energies $E_{l\boldsymbol{q}}$. In order to calculate $\varepsilon_M(\boldsymbol{k},\omega)$ in the SCF, the equation corresponding to (41) is

$$n(\boldsymbol{k}+\boldsymbol{G},\omega)=\sum_{\boldsymbol{G}'}\chi^0(\boldsymbol{k}+\boldsymbol{G},\boldsymbol{k}+\boldsymbol{G}';\omega)(-e)\varphi_{tot}(\boldsymbol{k}+\boldsymbol{G}',\omega), \quad (57)$$

where now

$$\varphi_{tot}(\boldsymbol{k}+\boldsymbol{G}',\omega)=\delta_{\boldsymbol{G}'\boldsymbol{0}}\varphi_{ext}(\boldsymbol{k},\omega)+\varphi_{ind}(\boldsymbol{k}+\boldsymbol{G}',\omega). \quad (58)$$

Within the RPA,

$$\varphi_{ind}(\boldsymbol{k}+\boldsymbol{G}',\omega)=-\frac{4\pi e}{(\boldsymbol{k}+\boldsymbol{G}')^2}n(\boldsymbol{k}+\boldsymbol{G}',\omega). \quad (59)$$

Using again (57) yields

$$\varphi_{ind}(\boldsymbol{k}+\boldsymbol{G}',\omega)=\frac{4\pi e^2}{(k+\boldsymbol{G}')^2}\sum_{\boldsymbol{G}''}\chi^0(\boldsymbol{k}+\boldsymbol{G}',\boldsymbol{k}+\boldsymbol{G}'';\omega)\varphi_{tot}(\boldsymbol{k}+\boldsymbol{G}'',\omega). \quad (60)$$

Combining (60) and (58), we obtain

$$\sum_{\boldsymbol{G}''}\left[\delta_{\boldsymbol{G}'\boldsymbol{G}''}-\frac{4\pi e^2}{(\boldsymbol{k}+\boldsymbol{G}')^2}\chi^0(\boldsymbol{k}+\boldsymbol{G}',\boldsymbol{k}+\boldsymbol{G}'';\omega)\right]\cdot\varphi_{tot}(\boldsymbol{k}+\boldsymbol{G}'',\omega)\equiv\sum_{\boldsymbol{G}''}\varepsilon_{\boldsymbol{G}'\boldsymbol{G}''}(\boldsymbol{k},\omega)\varphi_{tot}(\boldsymbol{k}+\boldsymbol{G}'';\omega)=\delta_{\boldsymbol{G}'\boldsymbol{0}}\varphi_{ext}(\boldsymbol{k},\omega),$$

where the *microscopic* dielectric matrix defined by

$$\varepsilon_{\boldsymbol{G}\boldsymbol{G}'}(\boldsymbol{k},\omega)=\delta_{\boldsymbol{G}\boldsymbol{G}'}-\frac{4\pi e^2}{(\boldsymbol{k}+\boldsymbol{G})^2}\chi^0(\boldsymbol{k}+\boldsymbol{G},\boldsymbol{k}+\boldsymbol{G}';\omega) \quad (61)$$

is introduced. If we define the inverse dielectric matrix $\varepsilon^{-1}_{\boldsymbol{G}\boldsymbol{G}'}(\boldsymbol{k},\omega)$ by

$$\sum_{\boldsymbol{G}'}\varepsilon^{-1}_{\boldsymbol{G}\boldsymbol{G}'}(\boldsymbol{k},\omega)\varepsilon_{\boldsymbol{G}'\boldsymbol{G}''}(\boldsymbol{k},\omega)=\delta_{\boldsymbol{G}\boldsymbol{G}''} \quad (62)$$

(note that $\varepsilon^{-1}_{\boldsymbol{G}\boldsymbol{G}'}(\boldsymbol{k},\omega)\neq(1/\varepsilon_{\boldsymbol{G}\boldsymbol{G}'}(\boldsymbol{k},\omega))$, then we can solve formally for $\varphi_{tot}(\boldsymbol{k}+\boldsymbol{G},\omega)$ and find

$$\varphi_{tot}(\boldsymbol{k}+\boldsymbol{G},\omega)=\varepsilon^{-1}_{\boldsymbol{G}\boldsymbol{0}}(\boldsymbol{k},\omega)\varphi_{ext}(\boldsymbol{k},\omega), \quad (63)$$

and finally for $\boldsymbol{G}=\boldsymbol{0}$ from (34)

$$\frac{1}{\varepsilon^{RPA}_M(\boldsymbol{k},\omega)}=\varepsilon^{-1}_{\boldsymbol{0}\boldsymbol{0}}(\boldsymbol{k},\omega) \quad (64)$$

or

$$\varepsilon^{RPA}_M(\boldsymbol{k},\omega)=1/\varepsilon^{-1}_{\boldsymbol{0}\boldsymbol{0}}(\boldsymbol{k},\omega)=\varepsilon_{\boldsymbol{0}\boldsymbol{0}}(\boldsymbol{k},\omega)-\sum_{\boldsymbol{G}\neq\boldsymbol{0},\boldsymbol{G}'\neq\boldsymbol{0}}\varepsilon_{\boldsymbol{0}\boldsymbol{G}}(\boldsymbol{k},\omega)M^{-1}_{\boldsymbol{G}\boldsymbol{G}'}(\boldsymbol{k},\omega)\varepsilon_{\boldsymbol{G}'\boldsymbol{0}}(\boldsymbol{k},\omega), \quad (65)$$

where M^{-1} is the inverse of the submatrix M which is the dielectric matrix ε without the first row and first column. The first term of the right-hand side of (65) describes the intra- and interband contributions to the dielectric function, the second term accounts for the so-called local field effect that arises from the (periodic) inhomogeneity of the crystalline solid.

More generally, we have for the dynamic structure factor of a crystalline solid (for $\hbar\omega\gg k_BT$) in the RPA

$$S(\boldsymbol{K},\omega)=-\frac{\hbar(\boldsymbol{k}+\boldsymbol{G})^2}{4\pi^2e^2n}\operatorname{Im}[\varepsilon^{-1}_{\boldsymbol{G}\boldsymbol{G}}(\boldsymbol{k},\omega)] \quad (66)$$

with

$$\boldsymbol{k}_0-\boldsymbol{k}_1=\boldsymbol{K}=\boldsymbol{k}+\boldsymbol{G}. \quad (67)$$

We now have to distinguish between the scattering vector $\boldsymbol{K}=\boldsymbol{k}_0-\boldsymbol{k}_1$ and the crystal momentum $\boldsymbol{k}$, which is restricted to the first BZ. In other words, in order to obtain the dynamic structure factor, we have to calculate the microscopic dielectric matrix, invert it and take the $\boldsymbol{G}\boldsymbol{G}$ element, where the appropriate $\boldsymbol{G}$ is determined by the requirement (67). For simple metals and semiconductors, the nearly-free-electron pseudopotential approximation can be applied to calculate the dielectric matrix by perturbation theory within a certain $(\boldsymbol{k},\omega)$ regime [17].

The dielectric matrix can be generalized to include xc-effects in some approximate way as in the density functional theory.

The above scheme, however, is not very practical when we deal with the response of tightly bound electrons as in insulators. These, to a good approximation, can be viewed as a collection of independent polarizable units with a local polarizability $\alpha(\omega)$. Including the intra-atomic Coulomb interaction, $\alpha(\omega)$ can be calculated self-consistently [26, 27] from the independent-particle polarizability

$$\alpha^0(\omega)=-\tfrac{1}{3}\int d^3r'\int d^3r\, r\,\chi^0_a(\boldsymbol{r},\boldsymbol{r}';\omega)\boldsymbol{r}'. \quad (68)$$

Here the index a denotes an atomic-like response function. Allowing the polarizable units to interact via

their dipolar fields leads to the well known Clausius-Mossotti formula in the long-wavelength limit or its generalization [28] to finite $\boldsymbol{k}$.

In metals it comes to an interplay of the core and conduction-electron responses that, treated in the SCF approximation [29, 30], provides, among other things, a simple explanation of the so-called soft-x-ray singularity observed in the core absorption spectra of simple metals [31].

I am grateful to R. O. Jones for a critical reading of the manuscript.

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