X-Ray Scattering from an Electron Gas*

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We show that the x-ray scattering from an electron gas in the large-momentum-transfer regime ($k \gg P_F/\hbar$) is a direct measure of the one-dimensional momentum distribution of the particles (including correlations). If the recoiling electron's momentum is measured in coincidence with the scattered x ray, then one can (in principle) measure the complete three-dimensional momentum distribution function.

I. CALCULATION

SUPPOSE we consider the incoherent scattering of x rays from an electron gas (see Fig. 1). If the frequency of the incoming x-ray beam is much higher than the plasma frequency, i.e., $\omega_1 \gg \omega_p$ and $E_F \ll \hbar\omega_1 < mc^2$, then the differential scattering cross section per unit volume of plasma is given by[1]

$$\frac{d\sigma}{d\omega d\Omega} = r_0^2 (\mathbf{e}_1 \cdot \mathbf{e}_2)^2 \sum_{\mathbf{p}} \sum_{\mathbf{p}'} \int_{-\infty}^{+\infty} \frac{dt}{(2\pi)} e^{-i\omega t} \times \langle a_{\mathbf{p}}^\dagger(t) a_{\mathbf{p}+\mathbf{k}}(t) a_{\mathbf{p}'+\mathbf{k}}^\dagger a_{\mathbf{p}'} \rangle, \quad (1)$$

where

$$\omega = \omega_1 - \omega_2 \quad \text{and} \quad \mathbf{k} = \mathbf{k}_1 - \mathbf{k}_2. \quad (2)$$

We use units in which $\hbar = c = 1$. Here $r_0 = e^2/m$ is the classical electron radius, $\mathbf{e}_1$ and $\mathbf{e}_2$ are the polarization vectors of the incoming and outgoing photon beams, respectively, and $a_{\mathbf{p}}$ is the annihilation operator for an electron in a state of momentum $\mathbf{p}$. The symbol $\langle\ \rangle$ represents for zero temperature the ground-state expectation value and for finite temperature the usual statistical average, and

$$a_{\mathbf{p}}(t) = e^{iHt} a_{\mathbf{p}} e^{-iHt}. \quad (3)$$

For zero temperature, the ground state of the interacting electron gas is completely specified by the momentum distribution $n_{\mathbf{p}}$.

$$n_{\mathbf{p}} = \langle a_{\mathbf{p}}^\dagger a_{\mathbf{p}} \rangle. \quad (4)$$

If we assume a noninteracting electron gas $n_{\mathbf{p}}$ is of course unity for $p < P_F$ (the Fermi momentum) and is equal to zero for $p > P_F$. In an interacting electron system collisions will produce a smearing of the distribution function.

Luttinger has shown[2] that even in the interacting system there is still a discontinuity in the distribution function at $p = P_F$. The exact magnitude of the discontinuity is unknown. However, for real metals it is expected to be of order unity. In addition, it is known[3] that a tail will develop on the distribution function. The tail is algebraic in character with a falloff in momentum space for large values of p which goes at least as fast as $(P_F/p)^6$.[4] In any case, it is safe to assume that $n_{\mathbf{p}}$ is only finite for values of p which are of the order of P_F. Everything we will prove about the exact scattering amplitude will be correct only to order (P_F/k). We will assume that $n_{\mathbf{p}+\mathbf{k}} = 0$. This introduces an error at most of order $(P_F/k)^6$. Errors of order $(P_F/k)^2$ will be introduced when at a later stage we neglect the energy of the particles in the Fermi sea relative to their recoil energy.

In Eq. (1) the operator $a_{\mathbf{p}'}$ operates directly (to the right) on the ground state of the system. The operator $a_{\mathbf{p}}^\dagger$ operates directly (to the left) on the ground state of the system. This implies that both p and p' are of the order of magnitude of P_F. By our assumption on the size of k, $|\mathbf{p}+\mathbf{k}| \gg P_F$ and $|\mathbf{p}'+\mathbf{k}| \gg P_F$. We will assume in rewriting the correlation function in Eq. (1) that

$$a_{\mathbf{p}+\mathbf{k}}(t) = a_{\mathbf{p}+\mathbf{k}} \exp -i\epsilon_{\mathbf{p}+\mathbf{k}} t, \quad (5)$$

i.e., that to a very good approximation the fast particle behaves like a free particle. Here $\epsilon_{\mathbf{p}+\mathbf{k}} \equiv (\mathbf{p}+\mathbf{k})^2/2m$.

In using Eq. (5) we have neglected the interaction between the fast particle (of momentum $\mathbf{p}+\mathbf{k}$) and the remaining particles in the medium. Interaction effects will, crudely speaking, introduce an imaginary part into the energy in Eq. (5) (lifetime effects), and produce a

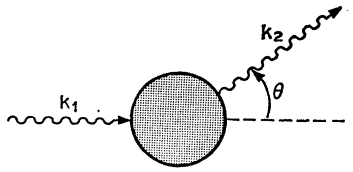

FIG. 1. Diagrammatic description of the incoherent photon scattering.

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[1] M. N. Rosenbluth and N. Rostoker, Phys. Fluids **5**, 776 (1962); D. F. Du Bois and V. Gilinsky, Phys. Rev. **133**, A1308 (1964).

[2] J. M. Luttinger, Phys. Rev. **119**, 1153 (1960); **121**, 942 (1961).

[3] See Ref. 2 and also E. Daniel and S. H. Vosko, Phys. Rev. **120**, 2041 (1960).

[4] This arises from the requirement that the average kinetic energy should be finite. In Ref. 3 it is shown that to the lowest order in a perturbation expansion of the distribution function $n(p) \underset{\sim}{\to} (P_F/p)^8$ for $p \gg P_F$.

shift in the energy $\epsilon_{\mathbf{p}+\mathbf{k}}$ (self-energy effects). In order to estimate the error introduced by this approximation we have to evaluate the effects of collisions on a fast particle of momentum $\mathbf{k}'$ ($k' \equiv |\mathbf{p}+\mathbf{k}| \gg P_F$).

In real metals the plasma frequency ω_p, the Fermi energy ϵ_F, and the average potential energy $\langle \text{P.E.} \rangle = e^2/a_0$ (a_0 is the interparticle radius) are of the same order. Using a simple calculation one finds that the time between collisions $t \cong 1/\omega_p (k/k_F)^3$. This indicates that the effect of collision on the spreading of the energy is

$$\text{Im}E_{\mathbf{k}'} \cong (\hbar\omega_p)(P_F/k')^3, \tag{6}$$

where $E_{\mathbf{k}'}$ is the renormalized energy of an electron with momentum $\mathbf{k}'$. A simple Hartree-Fock calculation shows that collision corrections introduce a shift in the energy of the order of

$$\Delta(\text{Re}E_{\mathbf{k}'}) \cong (\hbar\omega_p)^2/\epsilon_{\mathbf{k}'}. \tag{7}$$

In addition to the self-energy effects considered here, one must also include the scattering of the fast electron from the "hole" in the Fermi sea. This effect produces corrections of the same order of magnitude as those taking into account the calculations of the $\text{Im}E_{\mathbf{k}'}$. This, crudely speaking, means that the electron-electron mean free path is of the same order as the electron-hole mean free path. We therefore conclude that our assumption that the recoil electron is not affected by collisions is a good approximation. Errors in the energy which decrease as k' increases may be neglected; only errors which are proportional to k' can spoil the conclusions reached here.

The fact that a high-energy particle behaves accurately like a free particle is a strict consequence of the *Coulomb interaction*. The conclusions reached here cannot be simply generalized to other laws of forces, particularly hard spheres. For hard spheres the collision time is inversely proportional to the momentum so that the $\text{Im}E_{\mathbf{k}'}$ is proportional to k'.

Using Eq. (5) we may write

$$\langle a_{\mathbf{p}}^\dagger(t) a_{\mathbf{p}+\mathbf{k}}(t) a_{\mathbf{p}'+\mathbf{k}}^\dagger a_{\mathbf{p}'} \rangle \cong \exp -i\epsilon_{\mathbf{p}+\mathbf{k}} t \times \langle a_{\mathbf{p}}^\dagger(t) a_{\mathbf{p}'} a_{\mathbf{p}+\mathbf{k}} a_{\mathbf{p}'+\mathbf{k}}^\dagger \rangle. \tag{8}$$

The operator $a_{\mathbf{p}'+\mathbf{k}}$ operating on the ground state of the interacting system creates, with probability one, an electron of momentum $\mathbf{p}'+\mathbf{k}$ since (by our assumption on the magnitude of $\mathbf{k}$), the state $\mathbf{p}'+\mathbf{k}$ is unoccupied. The operator $a_{\mathbf{p}+\mathbf{k}}$ must annihilate with unit probability this high-energy particle so that the matrix element vanishes unless $\mathbf{p}' = \mathbf{p}$. We finally obtain

$$\langle a_{\mathbf{p}}^\dagger(t) a_{\mathbf{p}+\mathbf{k}}(t) a_{\mathbf{p}'+\mathbf{k}}^\dagger a_{\mathbf{p}'} \rangle = \langle a_{\mathbf{p}}^\dagger(t) a_{\mathbf{p}} \rangle \exp -i\epsilon_{\mathbf{p}+\mathbf{k}} t. \tag{9}$$

Substituting Eq. (9) into Eq. (1) we obtain the cross section

$$\frac{d\sigma}{d\omega d\Omega} = r_0^2 (\mathbf{e}_1 \cdot \mathbf{e}_2)^2 \sum_{\mathbf{p}} \int_{-\infty}^{+\infty} \frac{dt}{2\pi} \times \langle a_{\mathbf{p}}^\dagger(t) a_{\mathbf{p}} \rangle \exp i(\omega - \epsilon_{\mathbf{p}+\mathbf{k}}) t. \tag{10}$$

Neglecting the time dependence of $\langle a_{\mathbf{p}}^\dagger(t) a_{\mathbf{p}} \rangle$ introduces an error of the order $p^2/2m$ relative to $k^2/2m$. To be consistent then we write

$$\epsilon_{\mathbf{p}+\mathbf{k}} = (k^2/2m) + (\mathbf{p}\cdot\mathbf{k}/m) \tag{11}$$

and obtain

$$\frac{d\sigma}{d\omega d\Omega} = \frac{r_0^2 (\mathbf{e}_1 \cdot \mathbf{e}_2)^2}{(2\pi)^3} \int \delta\left(\omega - \frac{k^2}{2m} - \frac{\mathbf{p}\cdot\mathbf{k}}{m}\right) n_{\mathbf{p}} d\mathbf{p}, \tag{12}$$

which is the desired result.

The procedure we have used here is exactly equivalent to the procedure used in evaluating the Compton scattering from bound electrons when the recoil energy is much larger than the binding energy.[5] One simply neglects all binding energies. Physically, our approximation rests on the following argument. The wavelength of the photon is so small (large k) that it interacts with an individual electron and ejects it from the Fermi sphere instantaneously (large recoil energy). The rest of the system, i.e., the remaining electrons and the hole are left to evolve by themselves via their total Hamiltonian (including correlations). This approximation is known in the literature as the "impulse approximation.[6]"

We wish to point out here that our result Eq. (12) can be obtained from Eq. (10) in a more formal way even for finite temperature, the only difference being that $n_{\mathbf{p}}$ will be the distribution function for finite temperature. Our starting point is Eq. (10) where $\langle\ \rangle$ now means the usual thermal average.

Using the spectral analysis method one may easily show that

$$\frac{d\sigma}{d\omega d\Omega} = r_0^2 (\mathbf{e}_1 \cdot \mathbf{e}_2)^2 \sum_{\mathbf{p}} \int_{-\infty}^{+\infty} d\omega' \delta(\omega - \epsilon_{\mathbf{p}+\mathbf{k}} + \omega' + \mu) \times -\frac{1}{\pi} \frac{1}{e^{\beta\omega'}+1} \text{Im}\tilde{M}_{\mathbf{p}}(\omega'), \tag{13}$$

where

$$\tilde{M}_{\mathbf{p}}(\omega) = M_{\mathbf{p}}(z_l \to \omega + i\eta), \tag{14}$$

$$M_{\mathbf{p}}(z_l) = \int_0^\beta du\, e^{uz_l} M_{\mathbf{p}}(u); \quad z_l = \frac{(2l+1)\pi i}{\beta}, \tag{15}$$

$$M_{\mathbf{p}}(u) = \langle a_{\mathbf{p}}^\dagger(u) a_{\mathbf{p}} \rangle, \tag{16}$$

and

$$a_{\mathbf{p}}(u) = e^{u(H-\mu N)} a_{\mathbf{p}} e^{-u(H-\mu N)}. \tag{17}$$

The function $\text{Im}\tilde{M}_{\mathbf{p}}(\omega')$ is finite for those values of $(\omega'+\mu)$ which correspond to the "dressed single-particle" excitation energies of the system.[7] We have assumed throughout that we can neglect the energy $(\omega'+\mu)$ relative to the recoil energy $\epsilon_{\mathbf{p}+\mathbf{k}}$ so that the argument of the delta function in Eq. (9) is independent

[5] J. W. Dumond, Rev. Mod. Phys. **5**, 1 (1933).
[6] G. F. Chew, Phys. Rev. **80**, 196 (1950).
[7] For a free electron gas $\text{Im}\tilde{M}_{\mathbf{p}}(\omega') = \pi\delta(\omega' - \epsilon_{\mathbf{p}} + \mu)$.

of ω'. The integral over ω' is easily performed, since

$$\frac{1}{\pi}\int \frac{d\omega'}{(e^{\beta\omega'}+1)} \mathrm{Im}\tilde{M}_p(\omega') \equiv n_p(T). \quad (18)$$

Equation (9) can now be generalized for finite temperatures by replacing n_p by $n_p(T)$. Here, the quantity $\beta=1/K_BT$ and μ is the chemical potential.

To put Eq. (9) in a more convenient form let us define the direction of the momentum transfer $\mathbf{k}$ as the z axis. Then the cross section takes the form

$$\frac{d\sigma}{d\omega d\Omega} = r_0^2(\mathbf{e}_1\cdot\mathbf{e}_2)^2\left(\frac{m}{k}\right)[f(p_z)]_{p_z=(\omega-k^2/2m)m/k}, \quad (19)$$

where

$$f(p_z) \equiv \int n_p dp_x dp_y/(2\pi)^3 \quad (20)$$

is the "one-dimensional distribution function." If we measure the direction of the recoiling electron, then it is easily shown that

$$\frac{d\sigma}{d\omega d\Omega_{\mathrm{ph}} d\Omega_{\mathrm{el}}} = \frac{r_0^2(\mathbf{e}_1\cdot\mathbf{e}_2)^2}{2\pi^2} n_p\left(\frac{m}{k}\right)(\mathbf{p}+\mathbf{k})^2, \quad (21)$$

where p is uniquely determined from the energy conservation condition:

$$\omega = k^2/2m + \mathbf{p}\cdot\mathbf{k}/m \quad (22)$$

since the direction of $\mathbf{p}+\mathbf{k}$ is fixed by the measurement of the recoiling electron.

II. DISCUSSION

Experimentally, it seems feasible to make a measurement of the kind considered here. For a 10-kV x ray, the recoiling electron will have an energy of the order of 400 eV for scattering in the backward direction. In general the recoil energy as a function of the scattering angle is given by

$$E_{\mathrm{recoil}} = [(\hbar\omega_1)^2/mc^2]\sin^2(\theta/2). \quad (23)$$

Typical Fermi energies for free electron-like metals are of the order of 5 eV so that we are indeed in the high-momentum-transfer regime. Since the cross section is of order 10^{-25} cm^2 per unit solid angle and there are roughly 10^{22} scatters per unit volume, one would expect one part in 10^3 of the incident beam to be scattered per cc of sample into a unit solid angle.

Scattering from the bound electrons, if the experiment were done in say an alkali metal, would produce background. The elastic scattering from bound electrons would not interfere with the highly inelastic free-electron scattering. The inelastic scattering from bound electrons, i.e., the ionization of the few outside electrons with the resulting scattering of the x rays will, in general, overlap with the incoherent scattering spectrum from the free electrons. However, one must, in scattering from bound electrons, supply the ionization energy plus recoil energy to the bound electrons. For potassium (K), this ionization energy is approximately 40 eV. For aluminum, it is approximately 75 eV. We arrange things experimentally so that the high-energy side (least energy loss, smallest ω) of the free-electron Compton line ($\omega_{\min}$) occurs at the smaller value of ω than the binding energy (E_B). This implies that there is a maximum recoil energy allowed for a fixed value of E_B and E_F, i.e.,

$$E_{\mathrm{recoil}} \leq E_B + 2E_F + [(E_B+2E_F)^2 - E_B^2]^{1/2}. \quad (24)$$

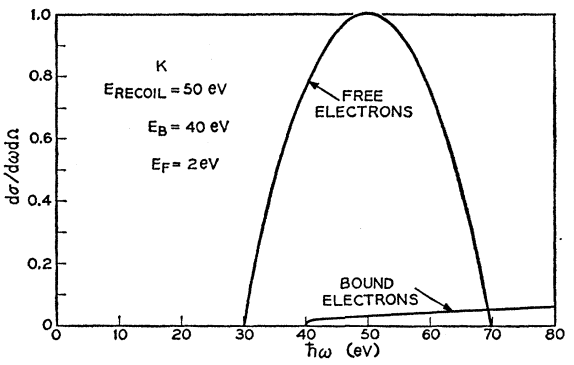

FIG. 2. The differential scattering cross section for free and bound electrons with a Fermi energy of 2 eV and a binding energy of 40 eV.

Correlation effects will produce a tail on the scattering in the region $\omega<\omega_{\min}$. They will also change the magnitude of the discontinuity in the slope of the curve at $\omega=\omega_{\min}$ and modify slightly the behavior of the cross section in the region $\omega>\omega_{\min}$. The bound electrons for the case $E_B>\omega_{\min}$ will *not interfere* with a measurement of the most interesting correlation effects: the existence of a tail and the change in the magnitude of the discontinuity at the "Fermi surface."

In Fig. 2 we have plotted the shape of the scattering cross section for a set of parameters pertinent to potassium. We have assumed that the electrons in the conduction band are free (noninteracting) with a Fermi energy of 2 eV, and that there is a single bound electron per atom having a hydrogenic wave function with a binding energy of 40 eV. The cross section is normalized in units of $(3/4n)(d\sigma/d\Omega)_{\mathrm{Th}}(1/kV_F)$, where $(d\sigma/d\Omega)_{\mathrm{Th}}$ is the single-electron Thomson cross section and n is the number of electrons per unit volume. In Appendix A we briefly outline the "approximate" calculation of the bound-electron piece. In the actual case we of course expect to see a tail in the region from $0<\omega<\omega_{\min}$, a change in the slope discontinuity and a modification of the line shape in the region $\omega>\omega_{\min}$.

Figure 3 is a similar plot for a series of parameters pertinent to Al (another free electron-like metal). The Fermi energy was taken to be 6 eV, and the binding

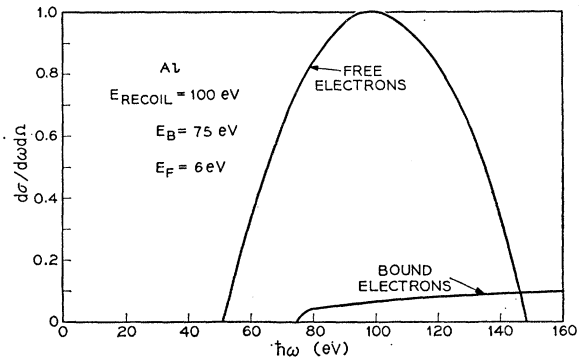

FIG. 3. The differential scattering cross section for free and bound electrons with a Fermi energy of 6 eV and a binding energy of 75 eV.

energy of the lowest bound electron was taken to be 75 eV.

In an actual experiment, core effects (the exclusion of the free electron from the central core) will produce high-momentum components in the wave function. When the noninteracting Fermi surface does not touch the zone boundary, then it is possible to show that these core effects do not interfere with the tail produced by correlation effects. When the noninteracting Fermi surface does overlap the zone boundaries, then core effects are mixed up with correlation effects. For the case of Al (or any multivalent free electron-like metal) the existence of a tail will not unambiguously determine the existence of correlation effects. We expect[8] that core effects in Al are approximately equal to or smaller than the effects of correlations so that it may still be possible to sort out the two effects.

It seems reasonable that an experiment can be done which would supply useful information about the interacting electron momentum distribution function. Positron annihilation data[9] give similar information. However, such data are complicated by electron-positron correlation effects occurring prior to their annihilation. If one assumes that the electron and the positron are uncorrelated (which is not true) then and only then will angular correlation measurement of the annihilation radiation lead to a measurement of the momentum distribution function.

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[8] W. A. Harrison, Phys. Rev. **131**, 2433 (1963).
[9] A. T. Stewart, J. H. Kusmiss, and R. H. March, Phys. Rev. **132**, 495 (1963).

APPENDIX

We will calculate the cross section for the incoherent scattering from a bound electron. It is approximately given by

$$\left(\frac{d\sigma}{d\omega d\Omega}\right)=\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{Th}}\sum_{\mathbf{q}}|\langle\mathbf{q}|e^{i\mathbf{k}\cdot\mathbf{x}}|\Phi\rangle|^2\delta(\epsilon_{\mathbf{q}}+E_B-\omega). \quad \text{(A1)}$$

Here $\epsilon_{\mathbf{q}}$ and $|\mathbf{q}\rangle$ are the energy and wave function, respectively, of the recoiling electron, k is the momentum transfer to system in the scattering process, and $|\Phi\rangle$ the ground-state wave function of the bound electron. If we assume that the final-state wave function is a plane wave then the cross section is given in terms of Φ_κ, the Fourier transform of $|\Phi\rangle$,

$$\left(\frac{d\sigma}{d\omega d\Omega}\right)_B=\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{Th}}\sum_{\kappa}|\Phi_\kappa|^2\delta(\epsilon_{\mathbf{q}+\kappa}+E_B-\omega). \quad \text{(A2)}$$

For simplicity, we shall assume that $|\Phi\rangle$ is hydrogenic, i.e.,

$$|\Phi\rangle=(1/(4\pi)^{1/2})(2/a^{3/2})e^{-r/a}, \quad \text{(A3)}$$

where a is the "Bohr radius" of the bound electron. We obtain for Φ_κ

$$\Phi_\kappa=8\sqrt{\pi}(l^{5/2})/(l^2+\kappa^2)^2, \quad \text{(A4)}$$

where $l=a^{-1}$. The cross section from the bound electron is given after single integration by

$$\left(\frac{d\sigma}{d\omega d\Omega}\right)_B=\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{Th}}\frac{8}{3\pi}\left(\frac{l^6}{qV_l}\right)\times\left\{\frac{1}{[\{(2m(\omega+E_B))^{1/2}-q\}^2+l^2]^3}-\frac{1}{[\{(2m(\omega+E_B))^{1/2}+q\}^2+l^2]^3}\right\}, \quad \text{(A5)}$$

for values of $\omega+E_B\geq 0$; where $V_l=l/m$. Simple calculations show that near the threshold, $\omega+E_B=0$, our formula for the cross section reduces to

$$\left(\frac{d\sigma}{d\omega d\Omega}\right)_B=\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{Th}}\frac{16}{\pi}\frac{1}{E_B}\frac{l^7}{(l^2+q^2)^4}(2m(\omega+E_B))^{1/2}. \quad \text{(A6)}$$

Equation (A6) was used in calculating Figs. 2 and 3.

Note added in proof. The main error made in doing this calculation is "neglecting" the time dependence of $\langle a_p^\dagger(t)a_p\rangle$. Actually, Eq. (12) would follow exactly from Eq. (10) if we had assumed that $a_p^\dagger(t)=\exp(ip^2/2m)a_p^\dagger(0)$. Although this is not correct, it is almost certainly approximately correct. A measure of the correctness is given for p near P_F by the ratio m^*/m for quasiparticles, which experimentally is about 1.25 for potassium.