Raman scattering of light by electrons in a metal with impurities

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The coefficient of reflection accompanied by a frequency change is calculated for a metal with impurities. It has a maximum at a frequency transfer determined by the condition that the excited-diffusion wavelength be equal to the electromagnetic-field penetration depth. The effect of anisotropy and of the electron interaction is considered.

Interest in Raman scattering of light by electrons has recently revived, since its observation permits the energy gap Δ to be determined, particularly in high-temperature superconductors, by a contactless method. In Refs. 1 and 2 we considered light scattering by superconductors with large as well as small correlation lengths $\xi_0 \sim v/\Delta$ (compared with the penetration depth δ of the light into the metal). The limiting case of a normal metal $\Delta \to 0$ was investigated in Ref. 3. This case is of interest also because, in view of the smallness of the scattering effect in superconductors, the corresponding value for a normal metal prior to the superconducting transition is usually subtracted in experiments.[4]

Raman scattering in pure metals was considered in all the preceding studies. Impurities, however, seem to play an important role in contemporary high-temperature superconductors. The present paper is devoted to the effect of impurities on Raman scattering in a normal metal.

As shown in Ref. 1, Raman scattering is determined by the Fourier component of the density correlator

$$f(q)=\int d^4x e^{-iq(x-y)}\langle\psi^+(x)\psi(x)\psi^+(y)\psi(y)\rangle,$$

where the angle brackets denote averaging over the ground state. In the case of a metal with impurities it is necessary, furthermore, to average over the impurity locations. To use for this purpose the known cross technique,[5] we relate, with the aid of the Landau theorem, the resultant correlator to retarded and advanced Green's functions:

$$\int dt e^{iq_0t}\langle\psi^+(x)\psi(x)\psi^+(y)\psi(y)\rangle$$
$$=-\int\frac{d\omega}{2\pi}\langle[G^R(x,y,\omega_+)-G^A(x,y,\omega_+)]$$
$$\times[G^R(y,x,\omega_-)-G^A(y,x,\omega_-)]\rangle\theta(\omega_+)\theta(-\omega_-),\tag{1}$$

where $t=x_0-y_0$ is the time difference, $x=(\mathbf{x},x_0)$, $\omega_\pm=\omega\pm q_0/2$. The remaining averaging is only over the impurities.

Since the impurity potential is independent of time, the frequency variable is conserved on a line that represents either of the Green's functions. In Fig. 1, for example, where the first impurity correction to the product $G^R G^A$ is shown, the upper line pertains only to G^R that correspond to the frequency ω_+ and there is no diagram with the product $G^R(\omega_+)G^A(\omega_+)$, while the lower pertains only to G^A with frequency ω_-; G_0 denotes Green's functions in the absence of impurities.

The equations that sum the influence of the impurities are similar to those obtained in a study of electric conductivity[5]:

$$\Pi^{RA}(p_+,p_-)=G^R(p_+)G^A(p_-)\Big[1+\frac{n}{(2\pi)^3}\int|u(\mathbf{p}-\mathbf{p}')|^2\Pi^{RA}$$
$$\times(p_+',p_-')d^3p'\Big],\tag{2}$$

where $p_+=(\mathbf{p}_+,\omega_+)$, while the Green's functions averaged over the impurities have, as is well known, the form

$$G^{R,A}(p)=(\omega-\xi\pm i/2\tau),\quad \frac{1}{\tau}=\frac{nmp_0}{(2\pi)^3}\int|u|^2d\Omega.$$

For a pointlike interaction with the impurity, $u(\mathbf{p}-\mathbf{p}')=u_0$, Eq. (2) can be easily solved. Introducing the notation

$$\Lambda^{RA}(\omega,\mathbf{q},q_0)=\frac{nu_0^2}{(2\pi)^3}\int\Pi^{RA}(p_+,p_-)d^3p$$

and integrating both sides of (2) with respect to p, we get

$$\Lambda^{RA}(\omega,\mathbf{q},q_0)=[1+\Lambda^{RA}(\omega,\mathbf{q},q_0)]I^{RA},\tag{3}$$

where

$$I^{RA}=\frac{nu_0^2}{(2\pi)^3}\int G^R(p_+)G^A(p_-)d^3p.$$

The integral with respect to the momentum can be represented, as usual, in the form of an integral with respect to the energy variable ξ and a variable s located on the equal-energy surface: $d^3p=d\xi ds/v$, where v is the velocity. In the final expressions $v=v_F$. Next, since we are interested in values of q and q_0 that are small compared with the Fermi momentum p_F and energy ε_F (the answer will contain a value of q not exceeding the reciprocal δ^{-1} of the light-penetration depth), we expand $\xi(p_\pm)$:

$$\xi(\mathbf{p}_\pm)=\xi(\mathbf{p})\pm\mathbf{vq}/2.$$

We obtain

$$I^{RA}=\frac{1}{8\pi^2\tau}\int\frac{d\xi\,d\Omega}{(\omega_+-\xi-\mathbf{vq}/2+i/2\tau)(\omega_--\xi+\mathbf{vq}/2-i/2\tau)}$$
$$=\frac{i}{4\pi\tau}\int\frac{d\Omega}{q_0-\mathbf{vq}+i/\tau}\tag{4}$$

It is seen from (4) that integration with respect to ξ causes

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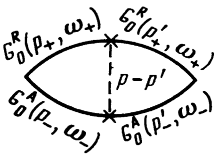

FIG. 1. First impurity correction to the electronic correlator.

I^{RR} and I^{AA} to vanish, and with them also Λ^{RR} and Λ^{AA}, since the poles of the corresponding Green's functions lie in this case in the same complex ξ half-plane.

The integral I^{RA} is easily calculated in two limiting cases—small and large q. For

$$|z|\ll 1, \text{ where } z=\frac{vq}{q_0+i/\tau}$$

we obtain accurate to terms quadratic in q

$$I^{RA}=\frac{i/\tau}{q_0+i/\tau}\left[1+\frac{1}{3}z^2\right]. \tag{5}$$

The Fourier component of the density correlator is expressed in terms of Π with the aid of (1):

$$f(q)=\frac{1}{(2\pi)^4}\int\limits_{-q_0/2}^{q_0/2}d\omega\int d\xi\frac{ds}{v}[\Pi^{RA}(\omega,\mathbf{q},q_0)+\Pi^{AR}(\omega,\mathbf{q},q_0)]$$
$$=\frac{q_0}{(2\pi)^4}\int d\xi\frac{ds}{v}\{G^R(p_+)G^A(p_-)[1-\Lambda^{RA}(\omega,\mathbf{q},q_0)]+\text{c.c.}\}.$$

After determining Λ from (3) and using the asymptote (5), we obtain for small q

$$f(q)=\frac{p_0mq_0}{2\pi^2}\left[\frac{i(1+z^2/3)}{q_0-iz^2/3\tau}+\text{c.c.}\right]. \tag{6}$$

It is clear from (6) why it was necessary to retain small terms of order q^2: the principal term, which does not contain q, is pure imaginary and does not contribute to the correlator. In addition, terms of order q^2, while small increments in the numerator of (6), are not small in the denominator at large $1/q_0\tau$, where a diffusion pole appears.

At large $|z|\gg 1$, the term with Λ in the correlator turns out to be a small correction of order $1/\tau vq$. Leaving it out, we obtain the value corresponding to the pure metal

$$f(q)=\frac{p_0mq_0}{(2\pi)^3}\int d\Omega\delta(\mathbf{vq}-q_0)=\frac{p_0mq_0}{(2\pi)^2vq}. \tag{7}$$

The scattering cross section is obtained by integrating, with respect to q, the density correlator with a factor that takes into account the electromagnetic-field distribution in the metal. Since greatest interest attaches to the optical frequency region of the incident radiation, and the frequency change is assumed small compared with the initial frequency, it can be assumed that the normal skin effect obtains for both the incident and scattered radiation. The factor describing the field distribution can then be written in the form[1]

$$\delta^{-3}(q^2+4/\delta^2)^{-2},$$

where δ is the field penetration depth at the incident-light frequency. In the integral with respect to q it is necessary to use for the density correlator either expression (6) or (7), depending on the region where the integrand is located.

In an extremely dirty metal, the electron mean free path is small compared with the skin-layer depth, i.e., $v\tau\ll\delta$. The main contribution to the integral comes from the region of small q (6), but the result depends also on the relation between the transferred frequency and $1/\tau$.

If $q_0\ll 1/\tau$, neglecting q_0 compared with i/τ, we need retain terms with q^2 only in the denominator of (6). We have then

$$f(q)=\frac{mp_0}{\pi^2}\frac{(q/q_d)^2}{1+(q/q_d)^4},$$

where $q_d=(3q_0/v^2\tau)^{1/2}$ is the value of the wave vector of the diffusion and corresponds to frequency transfer q_0. The scattering cross section $d\sigma$ is proportional to the integral

$$S=\frac{1}{\delta^3}\int\limits_0^{1/v\tau}\frac{dq}{(q^2+4/\delta^2)^2}\frac{(q/q_d)^2}{1+(q/q_d)^4}, \tag{8}$$

where the upper limit can be extended to infinity The cross section has thus a maximum at the transferred frequency determined from the condition that the wavelength of the excited diffusion be of the order of the skin layer $q_d\sim 2/\delta$ (see Fig. 2). On the two sides of the maximum, S is given by

$$S=\begin{cases}\dfrac{\pi}{2^{15/2}}\delta q_d, & q_d\ll 2/\delta, \qquad (9)\\[2ex] \dfrac{\pi}{8\delta^2q_d^2}, & (v\tau)^{-1}\gg q_d\gg 2/\delta. \qquad (10)\end{cases}$$

The upper bound on the diffusion wave vector is due to our previously assumed condition $q_0\tau\ll 1$.

At large transfers we have $q_0\gg 1/\tau$. The terms with q^2, in both the numerator and denominator of (6), are small increments that should be retained simultaneously. Expanding (6) in terms of q^2, we obtain

$$f(q)=p_0m(vq)^2/3\pi^2\tau q_0^3.$$

The cross section is proportional in this case to the integral

$$S=\frac{v^2}{3\tau\delta^3q_0^3}\int\limits_0^\infty\frac{q^2\,dq}{(q^2+4/\delta^2)^2}=\frac{\pi v^2}{24\tau\delta^2q_0^3}. \tag{11}$$

In investigations of light scattering in a pure metal with large mean free path, $v\tau\gg\delta$, it is also necessary to distinguish between small and large frequency transfers. The result for $q_0\gg 1/\tau$ was obtained in Refs. 1 and 3. We arrive at this result by using Eq. (7) for the density correlator. The cross section is then proportional to the integral

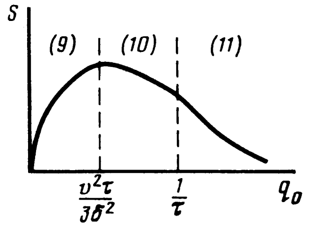

FIG. 2. Scattering cross section vs the transferred frequency for a dirty metal ($v\tau\ll\delta$); the numbers in the parentheses correspond to the numbers of the equations in the text.

$$S=\frac{q_0}{4v\delta^3}\int\limits_{q_0/v}^{\infty}\frac{dq}{q(q^2+4/\delta^2)^2}, \tag{12}$$

which has the following asymptotes

$$S=\begin{cases}\dfrac{1}{16}\left(\dfrac{v}{\delta q_0}\right)^3, & q_0\gg v/\delta, \qquad (13)\\[2ex] \dfrac{q_0\delta}{2^6 v}\ln\dfrac{2v}{q_0\delta}, & 1/\tau\ll q_0\ll v/\delta. \qquad (14)\end{cases}$$

At small frequency transfers, however, account must be taken of the contributions of both small and large wave vectors. Small q give rise to a contribution determined by Eqs. (8) and (9). The contribution of large q is given by the integral (12), whose lower limit is obtained from the condition that the short-wave asymptote be applicable (Eq. 7), i.e., it is now equal to $1/\tau v$. Noting that in this case $\delta\ll\tau v$, we obtain for this contribution

$$\frac{q_0\delta}{2^6 v}\ln\frac{2\tau v}{\delta}.$$

Joining the two contributions, we obtain the cross section in the region $q_0\ll 1/\tau$ for the considered case of a pure metal $\tau v\gg\delta$:

$$S=\frac{\delta}{2^6}\left(2^{1/2}\pi q_d+\frac{q_0}{v}\ln\frac{2\tau v}{\delta}\right). \tag{15}$$

The prefactor of the logarithm is here smaller than the first term, but the large value of the logarithm can offset this smallness. The dependences (13)–(15) of the scattering cross section on q_0 for a pure metal are shown in Fig. 3.

We present finally the connection between the value of S calculated above and the reflection coefficient $d\sigma$, which represents the fraction of incident photons reflected in a frequency interval $d\omega'$ into a solid angle $d\Omega'$:

$$d\sigma=\frac{2^8}{\pi^3}\left(\frac{\alpha p_0}{mc}\right)^2 P(n,\varkappa)S\frac{\delta}{v}d\omega'\,d\Omega', \tag{16}$$

where $\alpha = 1/137$ is the fine-structure constant,

$$P(n,\varkappa)=[(n+1)^2+\varkappa^2]^{-1}[(n-1)^2+\varkappa^2]^{-1}\cos^2\varphi,$$

n and $\varkappa$ are the refractive index and the extinction coefficient at the frequency ω, and φ is the angle between the polarization planes of the incident and reflected waves, assumed to propagate in a direction normal to the surface.

We have so far paid no attention to the question of fundamental character—the role of electron interaction. This interaction is known to suppress the density fluctuations. However, as shown in Ref. 6 for Raman scattering, the effect preserves the anisotropy: the scattering cross section (16) contains in lieu of $1/m^2$ the quantity

$$\overline{(m_{ik}^{-1}-\overline{m_{ik}^{-1}})^2},$$

where m_{ik}^{-1} is the reciprocal-effective-mass tensor, the short bar denotes averaging over the Fermi surface, and the long one averaging over that strip on the Fermi surface in which the velocity component normal to the surface vanishes.

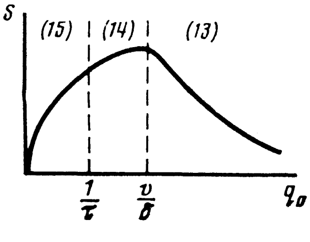

FIG. 3. Dependence of scattering cross section for a pure metal ($v\tau\gg\delta$).

In Ref. 6 was considered a pure superconductor, and its results apply to a pure normal metal, i.e., to Eqs. (13) and (14). If, however, the long-wave asymptote of the density correlator is used, it is necessary to take into account also the influence of the impurities on the electron interaction. This is in essence the question of Coulomb screening in a dirty metal. An answer to it is given in Ref. 7. The dielectric constant is in this case $\varepsilon(q_0,q) = 1 - \Pi(q_0,q)V(q)$, where $V(q) = 4\pi e^2/q^2$ is the Coulomb potential, $\Pi(q_0,q)$ is the electron polarization operator

$$\Pi(q_0,q) = -\frac{\varkappa_D^2}{4\pi e^2}\frac{Dq^2}{Dq^2-iq_0},$$

$\varkappa_D$ is the reciprocal Debye radius, and D is the diffusion coefficient. In the anisotropic case the product Dq^2 should be replaced by $D_{ik}q_iq_k$.

Repeating the calculations of Ref. 6, we find that the reciprocal effective mass $m_{ik}{}^{-1}A_{1i}A_{2k}$, which enters in the interaction with the magnetic field is replaced in the case of a dirty metal as follows:

$$m_{ik}^{-1}\to m_{ik}^{-1}+\overline{m_{ik}^{-1}}\Pi(q_0,q)V(q)/[1-\Pi(q_0,q)V(q)].$$

Substituting here the equations given above for $V(q)$ and $\Pi(q_0,q)$, we see that for the small frequency and wave-vector transfers of interest to us, meaning that so long as $\varkappa_D\gg\delta^{-1}$ and $\varkappa_D\gg q_d$ the replacement $m_{ik}{}^{-1}\to m_{ik}{}^{-1}-\overline{m_{ik}{}^{-1}}$ takes place in the vertex. Thus, the expression for the scattering cross section is subject to the same replacement as in the case of a pure metal, except that all the averagings of the quantity

$$\mu^2=\overline{(m_{ik}^{-1}-\overline{m_{ik}^{-1}})^2}$$

are carried out on the entire Fermi surface.

[1] A. A. Abrikosov and L. A. Fal'kovskiĭ, Zh. Eksp. Teor. Fiz. **40**, 262 (1961) [Sov. Phys. JETP **13**, 179 (1961)].

[2] A. A. Abrikosov and L. A. Falkovsky, Physica **C156**, 1 (1988).

[3] I. P. Ipatova, M. I. Kaganov, and A. V. Subashiev, Zh. Eksp. Teor. Fiz. **84**, 1830 (1983) [Sov. Phys. JETP **57**, 1066 (1983)].

[4] A. V. Bazhenov, A. V. Gorbunov, N. V. Klassen, *et al.*, Pis'ma Zh. Eksp. Teor. Fiz. **46**, Supplement, p. 35 (1987) [JETP Lett. **46**, Supplement, S29 (1987)].

[5] A. A. Abrikosov, L. P. Gor'kov, and I. E. Dzyaloshinskii, *Quantum Field-Theoretical Methods in Statistical Physics*, Pergamon, 1965 (p. 431 of Russ. original).

[6] A. A. Abrikosov and V. M. Genkin, Zh. Eksp. Teor. Fiz. **65**, 842 (1973) [Sov. Phys. JETP **38**, 417 (1974)].

[7] B. L. Al'tshuler and A. G. Aronov, *ibid.* **77**, 2028 (1979) [**50**, 968 (1979)].

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