Effect of a magnetic field on the Raman spectrum in a metal

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Inelastic scattering of light in a magnetic field is studied theoretically from the standpoint of determining the nature of the Raman spectra of high-T_c superconductors. A quasiclassical approach based on Boltzmann's equation is used. Two limiting cases are analyzed in detail — pure and dirty metals. In a pure crystal logarithmic peaks corresponding to cyclotron resonances should be observed. In the dirty case a wide relaxation continuum, which is determined by the reciprocal of the electron relaxation time, splits into smaller continua whose peaks lie near the transferred frequencies which are multiples of the cyclotron frequency. The skin effect and the Coulomb interaction are taken into account. Raman scattering with excitation of an optical phonon is analyzed. It is shown that in a magnetic field the phonon peak is broadened as a result of stronger phonon damping in the case where the frequency of the phonon lies near the cyclotron resonance.

1. Raman scattering of light is widely employed for investigating the spectra of different quasiparticles in crystals. The theory of Raman scattering for insulators has been well developed and agrees with the experimental data (see the review in Ref. 1). The existing theories cannot, however, explain recent experiments on the scattering of light in high-T_c superconductors.[2–7]. Most surprising is the behavior of the scattering cross section at high transferred frequencies. The cross section is virtually constant up to very high transferred frequencies, ~ 1 eV. It has been suggested[7] that because it is insensitive to the carrier density, this continuum is not associated with the conduction electrons. At the same time, the clearly visible Fano antiresonances, which correspond to phonon creation, in the spectral lines point to a process in which conduction electrons do participate. The existing theories, however, mainly describe electronic Raman scattering. It is of special interest, therefore, to find methods that would shed light on the origin on the Raman spectra of high-T_c superconductors. One such method could be the scattering of light in a magnetic field. The present calculation was undertaken for the purpose of obtaining a general picture of the phenomenon and to predict the form of the cross section. The scattering mechanism is assumed to be electronic.

2. It is convenient to study the electronic inelastic scattering of light[8,9] as a scattering process in an effective external field $U(\mathbf{r},t)$ which is bilinear in the vector potentials of the incident (i) and scattered (s) waves:

$$A^{(i)}(\mathbf{r},t)A^{(s)}(\mathbf{r},t) \simeq U(\mathbf{r},t)=U(z)\exp[i(\mathbf{k}_s\mathbf{s}-\omega t)], \tag{1}$$

where the transferred frequency $\omega=\omega^{(i)}-\omega^{(s)}$ and the transferred wave vector along the surface $\mathbf{k}_s=\mathbf{k}_s^{(i)}-\mathbf{k}_s^{(s)}$ are introduced. Here s is the coordinate vector along the surface; $\omega^{(i)}$, $\omega^{(s)}$ and $\mathbf{k}_s^{(i)}, \mathbf{k}_s^{(s)}$ are, respectively, the frequencies and wave vectors of the incident and scattered waves. The surface is located at $z=0$ and the crystal occupies the half-space $z>0$. The factor $U(z)$ describes the propagation of the field inside the crystal. In the case of a normal skin effect it has the form

$$U(z)=\exp(i\zeta z)=\exp(i\zeta_1 z-\zeta_2 z), \tag{2}$$

where ζ is the sum of the z-components of the wave vectors of the incident and scattered waves in the medium. The interaction of the field (2) with the electrons is described by the Hamiltonian

$$H_{\text{eff}}=\frac{e^2}{mc^2}\int \frac{d^3p}{(2\pi)^3}\gamma(\mathbf{p})U(\mathbf{r},t)f_p(\mathbf{r},t), \tag{3}$$

where $f_p(\mathbf{r},t)$ is the electron distribution function. The vertex function $\gamma(\mathbf{p})$ takes into account the polarization of light and the virtual interband transitions. The microscopic expression of the vertex function is not important in our case (see Ref. 10). Expression (3) assumes explicitly that the light-scattering process is quasiclassical. This is valid if the transferred frequencies and the wave vector of the light are small compared to the Fermi energy and Fermi momentum.

The scattering cross section calculated from the Hamiltonian (3) has the form[11]

$$\frac{d^2\sigma}{d\omega do}\simeq\frac{e^4}{m^2c^4}\frac{\Sigma(\mathbf{k}_s,\omega)}{1-\exp(-\omega/T)}, \tag{4}$$

where the correlation function $\Sigma(\mathbf{k}_s,\omega)$ can be expressed by means of the fluctuation-dissipation theorem in terms of the imaginary part of the generalized susceptibility of a system of electrons in the field $U(z)$:[8]

$$\Sigma(\mathbf{k}_s,\omega)=-2\,\text{Im}\int_0^\infty dz\; U^*(z)\int \frac{2d^3p}{(2\pi)^3}\gamma^*(\mathbf{p})f_p(\mathbf{k}_s,z,\omega), \tag{5}$$

where $f_p(\mathbf{k}_s,z,\omega)$ is the Fourier transform of the distribution function with respect to the time and coordinates in the plane of the surface.

The Boltzmann equation must therefore be solved for the electron distribution function in a magnetic field in order to find the cross section. We shall consider the case of normal incidence and scattering ($\mathbf{k_s}=0$). For simplicity we assume that the magnetic field is applied perpendicular to the surface (along the z axis). Instead of p_x and p_y, we employ as the variables the electron energy ε and the angle ϕ which denotes the position of an electron on a quasiclassical trajectory $\gamma(\mathbf{p})\rightarrow\gamma(p_z,\phi,\varepsilon)$, $d^3p\rightarrow m^*(p_z)dp_z\,d\phi\,d\varepsilon$, and $m^*(p_z)$ is the cyclotron mass. We seek the distribution function in the form

$$f_p(z,\omega)=f_0[\varepsilon_0+\gamma(p_z,\phi)U(z)-\mu]+\frac{df_0}{d\varepsilon}\chi_p(z,\omega). \tag{6}$$

The first term in this expression gives zero after substitution into the collision integral. For the boundary condition we choose the condition of specular reflection of electrons from the surface. A different choice of the boundary condition will not change the qualitative result. In order to use the Fourier transform in all space, we make an even continuation of $\chi_p(z,\omega)$ and $U(z)$ into the region $z<0$. Then the kinetic equation in the τ approximation assumes the standard form. We seek its solution in the form

$$\chi_p(k,\omega)=-\sum_n e^{in\phi}\frac{\omega\gamma^{(n)}(p_z)U(k)}{\omega-n\omega_H(p_z)-v_z(p_z)k+i\tau_p^{-1}}, \tag{7}$$

where $\omega_H(p_z)=eH/cm^*(p_z)$ is the cyclotron frequency. We introduce the following notation for the harmonics of the vertex function:

$$\gamma^{(n)}(p_z)=\int_0^{2\pi}\frac{d\phi}{2\pi}\gamma(p_z,\phi)e^{-in\phi}. \tag{8}$$

In the solution (7) v_z is assumed to be independent of ϕ (since $vk\ll\tau^{-1}$, this is not a serious limitation for high-T_c superconductors). It is easy to see that since $U(k)$ is even, solution (7) satisfies the chosen boundary condition.

To evaluate the effects of the electromagnetic field, we must take into account the term $\mathbf{v}\cdot\mathbf{E}$ in Eq. (7). These effects will be briefly discussed in Sec. 4.

Substituting expression (7) into (6) and then into (5), we obtain the cross section for electronic Raman scattering in a magnetic field

$$\Sigma(\omega)=-\mathrm{Im}\int\frac{dk}{2\pi}|U(k)|^2\sum_n\left\langle\frac{\omega|\gamma^{(n)}(p_z)|^2}{\omega-n\omega_H(p_z)-v_z(p_z)k+i\tau_p^{-1}}\right\rangle, \tag{9}$$

where the brackets designate an integral over the Fermi surface

$$\langle\ldots\rangle=\frac{2}{(2\pi)^2}\int dp_z\, m^*(p_z)(\ldots).$$

We now examine the result obtained in two limiting cases — a pure metal and a dirty metal. This can be done similarly to the manner in which the conductivity in a magnetic field is calculated.[12]

3. Expression (9) contains cyclotron resonances at the frequencies $\omega=n\omega_H(p_z)+v_z(p_z)k$. In a pure metal $|\zeta|v\gg\tau^{-1}$ an imaginary part appears in Eq. (9) as a result of circumscribing the pole. In the case where $\partial/\partial p_z(n\omega_H(p_z)+v_z(p_z)k)\neq 0$ everywhere, we have the case of a reference point. The neighborhood of the point $k_0=|(\omega-n\omega_H(p_z))/v_z(p_z)|_{\min}$ in this case makes the main contribution to the integral over k. The cross section is determined to logarithmic accuracy by the strip $(p_z^{(1)})$ on the Fermi surface, where the function $|(\omega-n\omega_H(p_z))/v_z(p_z)|$ assumes the minimum value:

$$\Sigma^{(n)}(\omega)\simeq\frac{\pi\omega}{v}\langle|\gamma^{(n)}(p_z^{(1)})|^2\rangle\begin{cases}\ln\left(\dfrac{|\zeta|}{\max(k_0,\tau^{-1}/v)}\right)/|\zeta|^2, & |\zeta|\gg k_0\\ |\zeta|^2/k_0^4, & |\zeta|\ll k_0.\end{cases} \tag{10}$$

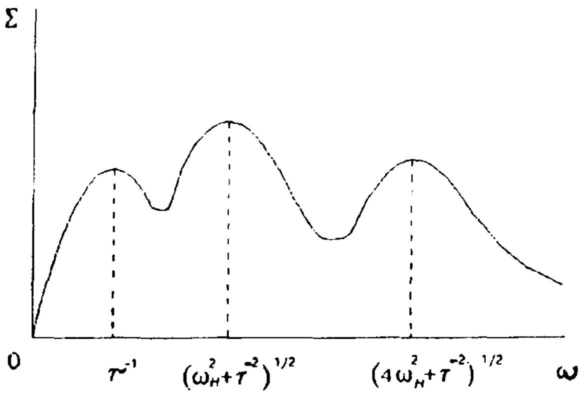

FIG. 1. Cross section for electronic Raman scattering in a magnetic field in a dirty crystal $v\zeta \ll \tau^{-1}$. When a magnetic field is switched on, the relaxation continuum with a maximum at $\omega = \tau^{-1}$ and width τ^{-1} splits into smaller continuua with maxima at $\omega = (n^2\omega_H^2 + \tau^{-2})^{1/2}$ and the same width. The ratio of the sizes of these continuua makes it possible to determine the symmetry of the effective electron-phonon vertex function.

In the case where at some point $p_z^{(2)}$ $\partial/\partial p_z(n\omega_H(p_z)+v_z(p_z)k)=0$, expanding in powers of p_z about this point up to second-order terms and extending the region of integration from $-\infty$ to $+\infty$, we obtain a lineshape corresponding to an asymmetric resonance. When $\zeta_1 \gg \zeta_2$ and $\tau^{-1} \gg v\zeta_2$ the lineshape has the form

$$\Sigma^{(n)}(\omega) \simeq \frac{\omega m^*(p_z^{(2)})|\gamma^{(n)}(p_z^{(2)})|^2}{2\pi\zeta_2|a_n|^{1/2}} \left(\frac{[(\omega-\Omega_n)^2+\tau^{-2}]^{1/2}-(\omega-\Omega_n)\operatorname{sgn} a_n}{(\omega-\Omega_n)^2+\tau^{-2}} \right)^{1/2}. \tag{11}$$

Here we introduce the notation

$$\Omega_n = n\omega_H(p_z^{(2)}) + v_z(p_z^{(2)})\zeta_1, \quad a_n = \frac{1}{2}\frac{d^2}{dp_z^2}[v_z(p_z)\zeta_1 + n\omega_H(p_z)]_{p_z=p_z^{(2)}},$$

where ζ_1 plays the role of k.

In the dirty case $|\zeta|v \ll \tau^{-1}$ (this situation is realized in high-T_c superconductors) the term vk in the denominator in Eq. (9) can be disregarded. As a result, we find

$$\Sigma(\omega) = \zeta_2^{-1}\sum_n \langle|\gamma^{(n)}(p_z)|^2\rangle \frac{\omega\tau^{-1}}{(\omega-n\omega_H)^2+\tau^{-2}}, \tag{12}$$

where for simplicity we set $\omega_H =$ const. The first term in this expression (with $n=0$) is a relaxation continuum, which is not affected by a magnetic field, with a maximum at $\omega \sim \tau^{-1}$ (Ref. 13). We see that a set of additional continua with peaks at $\omega = (n^2\omega^2 + \tau^{-2})^{1/2}$ appears in a magnetic field. These continua represent scattering by oscillators with characteristic frequencies $n\omega_H$ and damping τ^{-1}. All continuua have the same width τ^{-1} which does not depend on n. Their size is determined by the corresponding harmonics of the vertex function $\langle|\gamma^{(n)}(p_z)|^2\rangle$. The frequencies of the peaks in the continua are resolved when the magnetic field is sufficiently strong $\omega_H > \tau^{-1}$. The picture that should be observed in this case is illustrated in Fig. 1. In the opposite limit $\omega_H < \tau^{-1}$ they "collapse" into the standard continuum which is determined by the full vertex function $\Sigma\langle|\gamma^{(n)}(p_z)|^2\rangle$.

We see from Eqs. (10)–(12) that measurement of the Raman spectra in a magnetic field can give not only the electron-photon vertex function $\gamma(\mathbf{p})$ (it can answer the question of the degree to which the scattering is "electronic") but it also determines its symmetry — the nth resonance increases with increasing nth harmonic.

4. Let us briefly discuss the role of the dropped term with the electric field in the kinetic equation. To take this term into account, we must use a Maxwell's equation. The results obtained are as follows. The longitudinal part of the electromagnetic field gives the Coulomb screening of the zeroth-order vertex of the harmonic according to

$$\gamma^{(0)}(p_z) \to \gamma^{(0)}(p_z) = \gamma^{(0)}(p_z) - \langle \gamma^{(0)}(p_z)\rangle / \langle 1 \rangle.$$

The higher order harmonics, however, are not affected at all by the Coulomb interaction.

The transverse part of the electromagnetic field leads to the existence in the Raman spectrum of a peak which is associated with the excitation (absorption) of a helicon.[14] The helicon peak should be clearly observable in a sufficiently strong magnetic field, $\omega_H \gg \tau^{-1}$. Unfortunately, since the electron collision frequency in high-T_c superconductors, according to different estimates,[15] is rather high, $\tau^{-1} \sim 10^{12} - 10^{13}\ \mathrm{s}^{-1}$, this circumstance requires the use of strong magnetic fields, which cannot be produced at present.

5. In conclusion, we shall examine the effect of a magnetic field on Raman scattering accompanied by the excitation of an optical phonon. To take into account lattice vibrations, we must introduce into the kinetic equation a term describing the electron-phonon interaction. For self-consistency, we must write a separate equation for the phonons. We present the final result without repeating the simple calculations, which are identical to the calculations performed previously for the case of zero magnetic field:[16]

$$\Sigma(\omega) = -\mathrm{Im} \int \frac{dk}{2\pi} |U(k)|^2 \left(\Pi_{\gamma^*\gamma}(k,\omega) - \frac{\Pi_{\gamma^*\xi}(k,\omega)\Pi_{\xi^*\gamma}(k,\omega)}{\rho(\omega^2 - \omega_0^2(k)) - \Pi_{\xi^*\xi}(k,\omega)} \right), \tag{13}$$

where $\omega_0(k)$ is the bare phonon spectrum, ρ is the reduced-mass density of the lattice, and $\Pi(k,\omega)$ is the electronic loop (polarization operator) with indices denoting vertices, for example,

$$\Pi_{\xi^*\xi}(k,\omega) = \sum_n \left\langle \frac{(v_z(p_z)k - i\tau_p^{-1})\xi^{(n)*}(p_z)\xi^{(n)}(p_z)}{\omega - n\omega_H(p_z) - v_z(p_z)k + i\tau_p^{-1}} \right\rangle, \tag{14}$$

where $\xi(\mathbf{p})$ is the constant of the optical deformation potential. Expression (13), together with the results described above (first term in parentheses), contains a resonance at the phonon frequency whose shift as a result of the electron-phonon interaction is determined by the real part of the polarization operator (14). Just as in the case of a zero magnetic field, it has an asymmetric shape characteristic for a Fano antiresonance.[17,18] The width of the corresponding peak is determined by the phonon damping Γ, given by the imaginary part of expression (14). In a pure metal $vk \gg \tau^{-1}$ a calculation of the imaginary part gives the damping

$$\Gamma^{(n)} \simeq g^2 \frac{\omega_D^2}{vk} \theta(vk - |\omega - n\omega_H|).$$

Here $\theta(x)$ is the Heavside step function, ω_D is the Debye frequency ($\omega_D \sim \omega_0$), and g is the dimensionless electron-phonon coupling constant ($\xi \sim g\varepsilon_F$). For small wave vectors $\omega_D \gg vk \gg \tau^{-1}$ this expression increases near a cyclotron resonance. The maximum damping in this case is determined by the collisions

$$\Gamma_{\max} \simeq g^2 \omega_D^2 \tau. \tag{15}$$

We note that when $\tau^{-1} \leq g\omega_D$ this expression is not small. In a dirty crystal $vk \ll \tau^{-1}$ the damping has the form

$$\Gamma = \frac{1}{\rho} \sum_n \langle |\xi^{(n)}(p_z)|^2 \rangle \frac{\omega \tau^{-1}}{(\omega - n\omega_H)^2 + \tau^{-2}}$$

with the same maximum value (15). Upon excitation of an optical phonon we should therefore observe a broadening of the peak (up to the point at which it disappears completely) in the electronic Raman scattering spectrum if the phonon frequency is a multiple of the cyclotron frequency in the magnetic field.

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