Electron scattering of light in superconductors

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Electron Raman scattering of light has been observed recently in many high-temperature superconductors. At temperatures below the critical value there is a clear reduction in the scattering cross section at low transferred frequencies. In contrast to the theoretical predictions, there is no threshold which could be linked to the gap in the electron spectrum. This behavior of the scattering, and particularly its dependence on the transferred frequency, can be explained if we assume that the gap vanishes along certain lines on the Fermi surface.

There have been recent reports of the observation of electron Raman scattering of light (ERSL) in high-temperature superconductors. Such experimental observations make it possible to determine the most important parameter of a superconductor, which is the energy gap 2Δ in the spectrum, and then the ratio $2\Delta/T_c$ can be used to make conjectures concerning the superconductivity mechanism.

The first investigations of ERSL were made at the time of development of the theory of superconductivity. The first experimental attempt to carry out such measurements was unsuccessful.¹ A theoretical calculation² showed that the sensitivity of the apparatus had to be increased considerably: it was necessary to observe about 10^{-11} - 10^{-12} photons of the total number incident on a metal (light of optical wavelengths of 1 mW power represents about 10^{15} photons/s). The apparatus should also have a high spectral resolution: in the case of a superconductor with a gap of 5 meV the shifted Stokes line can be expected at 35 cm⁻¹ from the unshifted position (1 cm⁻¹ = 1.44 K).

The ERSL in superconductors was observed for the first time in the eighties.³⁻⁵ Comparison with the situation now encountered in high-temperature superconductors can be made on the basis of the results in Fig. 1 showing the dependence of the intensity of ERSL on the transferred frequency or frequency shift $\omega = \omega_i - \omega_s$ (ω_i and ω_s are the frequencies of the incident and scattered light) in the case of V₃Si at 5.48 K (Ref. 4). The critical temperature of the superconductor was $T_c = 18$ K. The peak at 42 cm⁻¹ in a section corresponding to about 8 photons/s shifted with temperature. The temperature in a laser spot formed on the surface of a superconductor was monitored. In the range 3.9-14.8 K the position of the peak agreed with the dependence $\Delta(T)$ deduced from the BCS theory. The ratio $2\Delta(0)/T_c$ was found to be 3.4, i.e., it was practically equal to the theoretical value 3.5. Similar results had been reported also for Nb₃Sn (Refs. 4 and 5).

It is worth noting particularly the profile of a peak (Fig. 1): it rises quite rapidly and shows a slow fall beyond the maximum. According to the theory of Refs. 2 and 6, the profile of the maximum depends on the relationship between the depth of penetration of light δ into a metal and the ratio v/Δ , where v is the characteristic value of the Fermi velocity. In the case of traditional superconductors, the ratio v/Δ gives the correlation length. If $v/\delta \gg \Delta$, we reach the limit of

large wave vectors transferred to a superconducting pair when light is scattered. The scattering has a threshold: it is not observed if the transferred frequency obeys $\omega < 2\Delta_{\min}$, where $2\Delta_{\min}$ is the minimum value of the gap in a Fermi surface belt where the component of the velocity normal to the surface of the metal vanishes. Above the threshold there is a fairly steep rise in the interval whose width is proportional to $\Delta \exp(-v/\delta \Delta)$ in the case of a weakly anisotropic gap, whereas it is $\Delta_{\max} - \Delta_{\min}$ in the case of a fairly strong anisotropy. On increase in the transferred frequency in the range $\omega - 2\Delta_{\max} \gtrsim \Delta$ the cross section reaches a value correspond-



FIG. 1. Raman spectra of V₃Si recorded at low temperatures.



FIG. 2. Raman spectrum of YBa₂Cu₃O_{7- δ} above the superconducting transition temperature.

ing to a normal metal. Therefore, strictly speaking, the position of the maximum cannot be used to find the gap. The minimum gap can be deduced from the value of the threshold. The difference between the position of the threshold and the maximum allows us to judge the anisotropy of the gap and it is clear from Fig. 1 that it amounts to 10%. This is an upper limit because the ratio is $v/\delta \sim 50 \text{ cm}^{-1}$ ($v \sim 10^8 \text{ cm/s}$, $\delta \sim 10^{-5} \text{ cm}$), i.e., it is of the same order of magnitude as Δ , so that we have a situation intermediate between large and small transferred vectors. The limit of small transferred vectors $v/\delta \ll \Delta$ applies to high-temperature superconductors if we bear in mind that the superconductivity is due to the BCS mechanism in the electron Fermi liquid.

Reports of the observation of ERSL in ceramic samples, films and single crystals of YBaCuO appeared in 1987–1988 (Refs. 7–11). Figures 2 and 3 are taken from Ref. 9 reporting work carried out on a YBa₂Cu₃O_{7- δ} single crystal at 3 and 90 K using two orientations—parallel (x, x) and crossed (x, y) polarizations: the first symbol gives the polarization of the incident light, the second that of the scattered light; light was incident along the c axis, the sample contained twins, and the symbols x and y represent the average spectra.



FIG. 3. Raman spectrum of $YBa_2Cu_3O_{7-\delta}$ at 3 K.

The sharp peaks in Figs. 2 and 3 describe the Raman scattering by phonons. In particular, the peaks at 115 and 330 cm^{-1} exhibit a strong temperature dependence and their profiles demonstrate that they represent what are known as the Fano resonances, i.e., they are due to phonon vibrations interacting strongly with conduction electrons.¹²

A comparison of Figs. 2 and 3 demonstrates that a wide continuum in the range 20–600 cm⁻¹ is very sensitive to the superconducting transition. If we estimate 2 Δ from the BCS relationship $2\Delta = 3.5T_c \approx 230$ cm⁻¹, we find that the continuum can describe ERSL.

Electron Raman scattering has been observed recently in superconducting bismuth¹³⁻¹⁶ and thallium^{17,18} systems. An estimate of the absolute cross section given in Ref. 17 is $(1-3) \times 10^{-12}$, which is in agreement with the theoretical calculation of Ref. 6 made on the assumption that the pairing mechanism is of the BCS type.

A characteristic feature of the electron spectra of all the currently known high-temperature superconductors is that the scattering occurs also in the range of small transferred energies 25-100 cm⁻¹, i.e., below the expected value of the gap in the spectrum. This can be explained by the presence of a certain amount of the normal phase in these samples.

The question now arises how ERSL should behave in the case of a normal metal. Unfortunately, this has not yet been investigated experimentally. According to the theory of Refs. 2 and 19, the scattering cross section of a normal metal has a fairly wide maximum at $\omega \sim v/\delta$ when the metal is sufficiently "pure." In the case of a "dirty" metal, i.e., when the depth of penetration of light δ is large compared with the mean free path of electrons l, the maximum is located at $\omega \sim lv/\delta^2$ (Ref. 20). The cross section falls to zero in the direction of lower transferred frequencies. Typical parameters of a high-temperature superconductor are $v \sim 0.5 \times 10^8$ cm/s, $\delta \sim 10^{-5}$ cm, and $v/\delta \sim 20$ cm⁻¹, whereas in the case of a "dirty" metal the position of the scattering maximum of the nonsuperconducting phase is shifted further toward even smaller transferred frequencies. Therefore, the experiments under discussion here were carried out in the range where there should be no fall as yet of the scattering cross section in the direction of smaller transferred frequencies. Such a fall was indeed absent at temperatures above the critical value.

At low temperatures the presence of a small amount of the normal phase should be manifested in the frequency range $v/\delta < \omega \ll \Delta$ as a term independent of the transferred frequency, as indeed was found for some samples. However, in the case of some other samples extrapolation of the cross section to $\omega = 0$ gives a fairly small value. Measurements of the specific heat²¹ of thallium samples grown by the same technology as the samples used to observe ERSL with a small cross section in the limit $\omega \rightarrow 0$ has failed to reveal a contribution linear in temperature in the superconducting region. It therefore follows that samples of this kind should not have a nonsuperconducting phase. There is however a fairly wide range $25 < \omega < 200$ cm⁻¹ where at low temperatures the ERSL process is characterized by an approximately linear dependence on the transferred frequency.

From the point of view of the theory of Ref. 6, in the limit of small transferred vectors $v/\delta \ll \Delta$, i.e., in the case of high-temperature superconductors, the ERSL spectrum has

a threshold at the minimum value of the gap for the Fermi surface. Therefore, the cross section should rise until the frequency exceeds the threshold by an amount Δ and it then should fall. In contrast to high-temperature superconductors, in this case a maximum of ERSL should be of the order of Δ irrespective of whether the gap is anisotropic. The cross section has no threshold if the minimum value of the gap vanishes. The cross section then rises quadratically with the frequency if the gap vanishes at isolated points on the Fermi surface and linearly if the gap vanishes along lines. If the gap vanishes somewhere on the Fermi surface, it is quite natural to expect that this occurs along lines in the case of layer superconductors, where the Fermi surface should have an almost cylindrical shape.

An analysis of electron Raman light scattering in hightemperature superconductors therefore leads to the following conclusions: 1) the minimum value in their case of the gap on the Fermi surface is small and possibly vanishes so that the ratio is $2\Delta_{\min}/T_c < 0.4$; 2) the gap reaches its minimum value along lines and not at individual points on the Fermi surface; 3) the maximum gap satisfies approximately the BCS relationship $2\Delta_{\max}/T_c \approx 4$.

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Translated by A. Tybulewicz