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Real Time Optical Spectroscopy of Superconducting-Gap Excitations

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Recently the interest in optical investigations of superconducting (SC) gaps increased /1 to 4/ in connection with the discovery of the high-temperature superconductivity and elucidating its nature. However, the results of these investigations often seem ambiguous. Thus, the SC gap shows up very weakly in the Raman scattering spectrum. It is possible to judge about its presence if one only takes the difference between spectra corresponding to the usual and SC phases /1, 2/. At the same time real time methods of vibrational spectroscopy have been developed /5 to 7/ due to the progress in generating ultrashort pulses (USP) of light. In the case of strongly damped motions these methods give information which cannot be obtained by usual frequency-domain spectroscopy /5/. Because of this, one can expect they will be also useful for superconductors.

In this work I suggest to use the methods of dynamical spectroscopy for the study of SC gaps. I have calculated the signal in the method of impulsive stimulated scattering /5/ when USP interact with the SC film. In this method the energy E_4 of the signal $\vec{k}_4$ generated due to the four-photon interaction of $\vec{k}_3 + \vec{k}_1 - \vec{k}_2$ type is measured. The beats in the dependence of energy E_4 on the delay time τ of probe USP $\vec{k}_3$ with respect to pump pulses $\vec{k}_1$ and $\vec{k}_2$ have been predicted. The beats are due to the vibrations of the charge density. Twice their period defines the value of the SC gap 2Δ. Consideration is made on the basis of the BCS phenomenological model /8/ for T = 0 K and the isotropic superconductor with large correlation length $\xi \gg \delta$. δ is the penetration depth of the electromagnetic field with vector potential $A_j(X)$; $X = \vec{r}, t$. The approximations similar to /3, 4/ were used.

The signal wave A_4 is generated by the non-linear current $j^{(3)}(X)$ in a medium which is produced by waves A_j (j = 1, 2, 3). We shall consider that pump and probe pulses do not overlap in time. Their durations $t_p \gg \omega^{-1}$, lc/n where $lc/n \sim \delta c/n$ is the propagation time of the field across a SC sample of thickness l. Because of this, the field amplitude A_j (j = 1, 2, 3) can be written as

$$A_j(\vec{r}, t) = (1/2)a_j^!(t)\exp\{-i[\omega t - \vec{k}_j(n + ib)\vec{r}]\} + \text{c.c.} \quad , \tag{1}$$

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where $|\vec{k}_j| \equiv k = \omega/c$. The value of current in a superconductor at given point $\vec{r}$ and given time moment t is determined by the average of the current operator in the secondary quantization representation:

$$j^{(3)}(X) = \frac{ie\hbar}{2m}\left\langle(\nabla\tilde{\Psi}^{+}_{\alpha})\tilde{\Psi}_{\alpha} - \tilde{\Psi}^{+}_{\alpha}(\nabla\tilde{\Psi}_{\alpha})\right\rangle - \frac{e^2}{mc}\left\langle\tilde{\Psi}^{+}_{\alpha}\tilde{\Psi}_{\alpha}\right\rangle A_3(X) \quad , \tag{2}$$

where $\tilde{\Psi}_{\alpha}^{+}$ and $\tilde{\Psi}_{\alpha}$ are the creation and destruction operators of a particle, respectively, in the Heisenberg representation. They depend on fields A_j. The summation is over spin indices α in (2). The further calculations show that the addend on the right-hand side of (2) gives the main contribution to $j^{(3)}$ for the case of light fields. One can show similarly to /3, 4/ that in the case under consideration the Hamiltonian of interaction with pump fields can be written as

$$H' = (e^2/mc^2)\Psi^{+}(X)\Psi(X)A_1(X)A_2(X) \quad . \tag{3}$$

The dependence of operators $\tilde{\Psi}^{+}$ and $\tilde{\Psi}$ on fields A_1 and A_2 can be obtained by the S-matrix:

$$\tilde{\Psi}(X) = S^{-1}(t)\Psi(X)S(t) \ .$$

The S-matrix obeys the equations

$$i\hbar\,\frac{\partial S}{\partial t} = H'S \ , \qquad i\hbar\,\frac{\partial S^{-1}}{\partial t} = -S^{-1}H' \ , \qquad S^{\pm 1}(-\infty) = 1 \ . \tag{4}$$

Calculating $j^{(3)}$ by (4) in a first approximation with respect to H' and using Wick's theorem, we obtain

$$j^{(3)}(X) = -i(2e^4/\hbar m^2c^3)\int_{-\infty}^{t} A_1(X')A_2(X')\{[-G(X' - X)G(X - X') - -F^2(X - X')] - \text{c.c.}\}d^4X'A(X) \quad , \tag{5}$$

where G(X) and F(X) are the normal and "abnormal" Green's function of a superconductor, respectively /9/. Expression (5) is calculated by the conversion to Fourier transforms of all quantities in the integrand. As a result, we obtain for the positive frequency component of the non-linear current

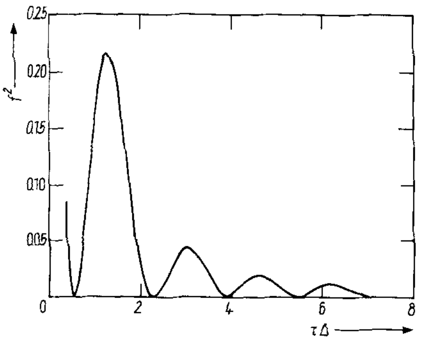
$$j^{(3)+}(\vec{r},\ t) = j^{(3)+}(t)L\exp\left[i\vec{k}_3\vec{r}(n + ib)\right], \tag{6}$$

where

$$j^{(3)+}(t) = \frac{e^4\Delta^2}{4(2\pi\hbar c)^3}\int_0^{\infty} ds\left[I_p(t - s) + I_p(t + s)\right]f(s\Delta)a'_3(t)\exp(-i\omega t) \tag{7}$$

defines the time dependence of current,

Fig. 1. Plot of f^2 versus $\tau\Delta$



$$I_p(t) = \mathrm{Re}\left[a_1'(t)a_2'^*(t)\right]/2 \quad ,$$

$$f(s\Delta) = J_o(s\Delta)Y_o(s\Delta) - J_1(s\Delta)Y_1(s\Delta) \quad ,$$

$J_n(s\Delta)$ and $Y_n(s\Delta)$ are the Bessel functions of the first and second kind, respectively. The following estimation holds for the factor L describing the non-locality of the interaction:

$$L \sim (kb)^{-1} \ln(2kbvt_p) \quad ;$$

v is the velocity on the Fermi surface.

Solving the Maxwell equations for the signal wave with current (6) we obtain the field amplitude of the signal wave after its passage across an SC sample

$$a_4(l,\ t) = -(4\pi L/c\kappa)j^{(3)+}(t)\left[l\ \exp(\kappa\ l) - \kappa^{-1}\ \mathrm{sh}(\kappa\ l)\right]$$

with

$$\kappa = -kb + ikn \quad . \tag{8}$$

In an experiment, it is convenient to record the energy of the signal $\vec{k}_4$

$$E_4(\tau) \approx \int dt\,|a_4(l,\ t)|^2$$

as a function of delay τ. In the case of sufficiently short both pump and probe pulses $t_p \ll \Delta^{-1}$, the time dependence $E_4(\tau)$ is determined by the function f: $E_4(\tau) \approx f^2(\tau\Delta)$. This function has the asymptotic representation

$$f(\tau\Delta) \approx -(2/\pi\tau\Delta)\cos(2\tau\Delta)$$

for large values of the argument. The dependence $f^2(\tau\Delta)$ appears in Fig. 1. The beats with frequency 4Δ are clearly seen there.

One can obtain the value of the ratio of signal and probe intensities by (7), (8). This value is

$$|a_4(l,\ t)|^2/|a_3^{!}(t)|^2 \approx 10^{-24}\ E_1E_2$$

for the characteristic magnitudes of parameters: $n \approx b \approx 3$, $l \approx 10^{-7}$ m, $\Delta/\omega \approx 10^{-3}$. Here $E_{1,2}$ are the energy densities of the pump pulses in terms of J/m^2. The possible values of $E_{1,2}$ are limited by the infinitesimal change of the quantities G and F, which characterize the SC state, during the time of pulse action, relative to their equilibrium values. This gives the upper bound of $E_j < 10^6\ J/m^2$. It should be noted that these values can be sufficient for heating-up the electron gas due to the absorption of radiation. As a result, the SC gap can diminish and even the SC state can be destroyed. However, in the case of sufficiently weak interaction between excitations the processes indicated correspond to the second, more slow stage of relaxation of the electron system of a superconductor to the equilibrium state /10/. These processes proceeds in times $\approx \Delta^{-1}$. Also, limitations on the pulse power are possible.

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Erratum

To the Short Note by B.D. FAINBERG

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the author requests the following corrections:

On p. K172 the numerical factor in the equation should read 10^{-15} instead of 10^{-24}.

On p. K172 in the 13th line from above in the sentence "These processes proceed in times $\approx \Delta^{-1}$" unfortunately some words have been omitted. The sentence reads correctly: "These processes do not present the observation of the effect predicted in the work, because it proceeds in times $\approx \Delta^{-1}$".

Besides, on p. K170 in formula (5) A(x) has to be replaced by $A_3(x)$ and in front of the right-hand side of formula (7) a minus sign has to be added.