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Inelastic scattering of light in superconducting Nb_3Sn†

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Theoretical and experimental results for the inelastic scattering of light off superconducting Nb_3Sn are compared. It is found that the experimental results by Fraas *et al.* support well the theoretical work by Cuden, discussing the scattering efficiency in terms of electronic intermediate interband transitions.

I. INTRODUCTION

Only recently the work by Fraas, Williams, and Porto[1] on photoluminescence studies of superconducting Nb_3Sn came to our attention. It is the purpose of this paper to compare their experimental results with the recent theoretical calculations by Cuden.[2]

In 1956 Khaikin and Bykov[3] tried to observe the energy gap in superconducting lead using light scattering techniques. However they were not able to observe the spatial distribution of the scattered light because of the small amount of scattering associated mainly with the fact that the skin effect allows the light to penetrate only into a very thin surface layer of a metal. The observation can be greatly impaired also by a background scattering and strong absorption.

Later, with the improved experimental technique Mooradian[4] observed that many metals exhibit a weak, broadband electronic luminescence when exposed to intense laser monochromatic light.

In 1970 Raman scattering experiments on tungsten bronzes were reported by Scott *et al.*[5] As pointed out by Tilley,[6] these authors studied scattering at frequency shifts above 200 cm^{-1} so that they probed the optical-phonon spectrum.

The frequency shifts of interest in superconductivity are of the order of the energy gap Δ, that is, a few wave numbers. In fact, the experiment by Fraas *et al.* has shown that in the superconducting Nb_3Sn scattering disappeared in the frequency region $\omega_{laser} > \omega > \omega_{laser} - \Delta$.

Furthermore their polarization measurements at 150 cm^{-1} indicated little intensity change with polarization indicating that the intermediate interband transitions are important.

In the theoretical work on Raman scattering of light off a superconductor by Abrikosov and Falkovskii,[7] Tong and Maradudin,[8] Tilley,[9] Dos Reis and Luzzi,[10] only intraband electronic intermediate transitions in a superconductor were considered. Their work exhibits a gap in the spectrum, but disagrees completely with the temperature and frequency dependence of the reflected light off a superconducting Nb_3Sn, found in the experiment by Fraas *et al.*

In a superconductor the vacuum-polarization processes dominate the long-wavelength polarizability of the electron gas because of the long-range Coulomb interaction and lead to effective screening of the BCS interaction. Dos Reis and Luzzi have considered the effects of screening on the scattering efficiency. In the static limit, however, their results reduce to the ones obtained by Abrikosov and Falkovskii with the reduced renormalized coupling constant.

In Ref. 2 the scattering efficiency was formulated using Kubo's nonlinear response theory in a form suitable for systematic diagrammatic expansion. Screening effects and the effects of the sample surface were taken into account. It was shown that the main contribution to the intensity of the inelastically scattered light comes from the unscreened fluctuations in transverse current, and not from density fluctuations as thought in earlier theories. The graphical representation of the four-photon Green's functions that describe the inelastic scattering of light by electronic excitations in a superconductor via intermediate intraband and interband transitions are shown in Figs. 1(a) and 1(b), respectively.

In this paper we present the inelastic scattering of light in a superconductor in terms of the conventional second-order time-dependent perturbation theory.[11] The emphasis is given to the simple physical picture of

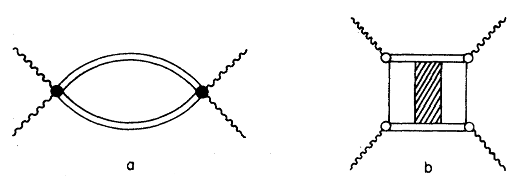

FIG. 1. Graphical representations of the four-vertex parts of the four-photon Green's functions that describe the inelastic scattering of light by one- and two-electron excitations in a superconductor.

the scattering processes involved. The fact that the electromagnetic fields seen by the electrons differ from the measured fields outside the metal is ignored.

II. HAMILTONIAN

Let us assume that the system is well represented by the Hamiltonian of the form

$$H=\sum_{\vec{p}\sigma}\epsilon_c(\vec{p})c^\dagger_{\vec{p}\sigma}c_{\vec{p}\sigma}-V\sum_{\vec{p}\vec{p}''}c^\dagger_{\vec{p}''\uparrow}c^\dagger_{-\vec{p}''\downarrow}c_{\vec{p}\downarrow}c_{-\vec{p}\downarrow}+\sum_{n\vec{p}\sigma}\epsilon_n(\vec{p})c^\dagger_{n\vec{p}\sigma}c_{n\vec{p}\sigma}\quad(n\neq c)\ , \tag{2.1}$$

$$\epsilon_c(\vec{p})=p^2/2m-\mu\ ,$$

where $\epsilon_c(\vec{p})$ is the kinetic energy of a conduction electron measured from the chemical potential μ, $\epsilon_n(\vec{p})$ is the energy of an electron in band n with wave vector $\vec{p}$, σ denotes the spin, and V is the effective BCS attractive interaction between the electrons with the opposite spins and momenta.

By the use of the well-known Bogoliubov-Valatin canonical transformation,

$$\begin{aligned}c^\dagger_{\vec{p}\uparrow}&=u_{\vec{p}}\gamma^\dagger_{\vec{p}\uparrow}+v_{\vec{p}}\gamma_{-\vec{p}\downarrow}R^\dagger\ ,\\ c_{\vec{p}\uparrow}&=u_{\vec{p}}\gamma_{\vec{p}\uparrow}+v_{\vec{p}}R\gamma^\dagger_{-\vec{p}\downarrow}\ ,\\ c^\dagger_{-\vec{p}\downarrow}&=u_{\vec{p}}\gamma^\dagger_{-\vec{p}\downarrow}-v_{\vec{p}}\gamma_{\vec{p}\uparrow}R^\dagger\ ,\\ c_{-\vec{p}\downarrow}&=u_{\vec{p}}\gamma_{-\vec{p}\downarrow}-v_{\vec{p}}R\gamma^\dagger_{\vec{p}\uparrow}\ ,\end{aligned} \tag{2.2}$$

the Hamiltonian (2.1) can be cast into the simple form

$$H=\sum_{\vec{p}\sigma}E_c(\vec{p})\gamma^\dagger_{\vec{p}\sigma}\gamma_{\vec{p}\sigma}+\sum_{n\vec{p}\sigma}\epsilon_n(\vec{p})c^\dagger_{n\vec{p}\sigma}c_{n\vec{p}\sigma}\ , \tag{2.3}$$

with

$$E_c(\vec{p})=(\epsilon_c^2(\vec{p})+\Delta^2)^{1/2}\ , \tag{2.4}$$

where $E_c(\vec{p})$ is the energy of the elementary excitations in a superconductor, and Δ is the temperature dependent order parameter. $u_{\vec{p}}$ and $v_{\vec{p}}$ are given by

$$u^2_{\vec{p}}=\frac{1}{2}\left[1+\frac{\epsilon_c(\vec{p})}{E_c(\vec{p})}\right]\ ,\quad v^2_{\vec{p}}=\frac{1}{2}\left[1-\frac{\epsilon_c(\vec{p})}{E_c(\vec{p})}\right]\ . \tag{2.5}$$

γ's and c's satisfy the usual Fermi anticommutation relations. The operators R^+ and R transform a given state in an N-particle system into the corresponding state in $(N+2)$- and $(N-2)$-particle systems, respectively.

The interaction between the electrons and the electromagnetic field represented by vector potential $\vec{A}(\vec{r},t)$ can be written

$$H'=H_A+H_{AA}\ , \tag{2.6}$$

$$H_A=\frac{e}{mc}\sum_{nn'\vec{k}\vec{q}\sigma}\langle n'\vec{k}+\vec{q}\sigma|e^{i\vec{q}\cdot\vec{r}}\vec{p}\cdot\vec{e}\,|n\vec{k}\sigma\rangle\times A(\vec{q},t)c^\dagger_{n'\vec{k}+\vec{q}\sigma}c_{n\vec{k}\sigma}\ , \tag{2.7}$$

$$H_{AA}=\frac{e^2}{2mc^2}\sum_{nn'\vec{k}\vec{q}_1\vec{q}_2\sigma}\langle n'\vec{k}+\vec{q}_1+\vec{q}_2\sigma|n\vec{k}\sigma\rangle\times\vec{A}(\vec{q}_1,t)\,\vec{A}(\vec{q}_2,t)c^\dagger_{n'\vec{k}+\vec{q}_1+\vec{q}_2\sigma}c_{n\vec{k}\sigma}\ . \tag{2.8}$$

(In Ref. 2 it was erroneously stated that the paramagnetic Hamiltonian H_A is a time-reversal-breaking Hamiltonian. Clearly, there are no spin-flip processes in H_A.)

Here $\vec{e}$ is a unit vector parallel to $\vec{A}(\vec{r},t)$ and $|n\vec{k}\sigma\rangle$ is the Bloch state vector. One obtains the long-wave-limit interaction Hamiltonian simply by taking the limit $\vec{q},\vec{q}_1,\vec{q}_2\rightarrow 0$ in Eqs. (2.7) and (2.8).

III. SCATTERING CROSS SECTION

The differential scattering cross section per unit frequency range per unit solid angle per unit volume of specimen for scattering of an incident photon with frequency ω_1, wave vector $\vec{q}_1$, and polarization $\vec{e}_1$, to a final photon with frequency ω_2, wave vector $\vec{q}_2$ and polarization $\vec{e}_2$ can be written as follows:

$$\frac{d^2\sigma}{d\Omega\,d\omega}=r_0^2\frac{\omega_2}{\omega_1}\sum_{if}|\langle f|t_{12}|i\rangle|^2(P_i-P_f)\times\delta(\omega-|E_f-E_i|)\ , \tag{3.1}$$

where within the framework of an independent quasiparticle model

$$\langle f|t_{12}|i\rangle=\vec{e}_1\cdot\vec{e}_2\delta_{f,i}+\frac{1}{m}\sum_l\left[\frac{\langle f|h_2|l\rangle\langle l|h_1|i\rangle}{E_i-E_l+\omega_1}+\frac{\langle f|h_1|l\rangle\langle l|h_2|i\rangle}{E_i+E_l-\omega_2}\right]\ , \tag{3.2}$$

$$h_1=\sum_{nn'\vec{k}\vec{q}\sigma}\langle n'\vec{k}+\vec{q}\sigma|e^{i\vec{q}\cdot\vec{r}}\vec{p}\cdot\vec{e}_1|n\vec{k}\sigma\rangle c^\dagger_{n'\vec{k}+\vec{q}\sigma}c_{n\vec{k}\sigma}\ , \tag{3.3}$$

$$h_2=\sum_{nn'\vec{k}\vec{q}\sigma}\langle n'\vec{k}+\vec{q}\sigma|e^{i\vec{q}\cdot\vec{r}}\vec{p}\cdot\vec{e}_2|n\vec{k}\sigma\rangle c^\dagger_{n'\vec{k}+\vec{q}\sigma}c_{n\vec{k}\sigma}\ , \tag{3.4}$$

$$r_0=e^2/mc^2\ ,\quad \omega=\omega_1-\omega_2\ ,\quad \vec{q}=\vec{q}_1-\vec{q}_2\ .$$

In above formulas r_0 is the Bohr radius of electron, ω is the transferred photon energy to the system, and $\vec{q}$ denotes the photon momentum transfer. Because of the low-temperature interval of interest ($T_c > T \geq 0$ °K) we consider in Eq. (3.1) the Stokes part of the spectrum only.

The summation in Eq. (3.1) is over the initial and final states, and in Eq. (3.2) over the intermediate states only. P_i and P_f are the thermal weight factors indicating the probability that at temperature T the initial and final states are occupied, respectively.

IV. SCATTERING OF LIGHT BY ONE-ELECTRON EXCITATIONS AT $T_c > T > 0$ °K

In what follows, we shall be primarily interested in the photon energy transfers in the range $0 < \omega \lesssim 6\Delta$, i.e., in the region of low-lying electronic excitations where $\omega = \omega_1 - \omega_2 << \omega_1, \omega_2$.

The "normal fluid" scattering cross section involves scattering of quasiparticles already present because of finite temperature. The excitation energy in this case can be arbitrarily small, as in the normal metal. The initial and final states are given by

$$|i\rangle = \gamma^\dagger_{\vec{p}\sigma}|0_s\rangle|0_n\rangle$$
$$|f\rangle = \gamma^\dagger_{\vec{p}+\vec{q}\sigma}|0_s\rangle|0_n\rangle \qquad (4.1)$$
$$\sigma = \uparrow, \downarrow$$

where the "vacuum states" $|0_s\rangle$ and $|0_n\rangle$ are defined by

$$\gamma_{\vec{p}\sigma}|0_s\rangle = 0 \ , \quad c_{n\vec{p}\sigma}|0_n\rangle = 0 \ , \quad \sigma = \uparrow, \downarrow \ . \qquad (4.2)$$

The corresponding initial- and final-state excitation energies are

$$E_i = E_c(\vec{p}) \ , \quad E_f = E_c(\vec{p}+\vec{q}) \ . \qquad (4.3)$$

The thermal-weight rate factor $(P_i - P_f)$ is in this case equal to

$$\begin{aligned} P_i - P_f &= f(E_c(\vec{p}))[1 - f(E_c(\vec{p}+\vec{q}))] \\ &\quad - f(E_c(\vec{p}+\vec{q}))[1 - f(E_c(\vec{p}))] \\ &= f(E_c(\vec{p})) - f(E_c(\vec{p}+\vec{q})) \ , \end{aligned} \qquad (4.4)$$

where $f(E)$ is the Fermi distribution function,

$$f(E) = 1/(e^{\beta E} + 1) \ . \qquad (4.5)$$

By using Eqs. (4.1), (3.3), and (3.4) the transition matrix element for the inelastic scattering ($i \neq f$) can be written

$$\begin{aligned} \langle f|t_{12}|i\rangle = \frac{1}{m} \sum_{n\vec{k}\delta m'\vec{p}''\vec{q}''\sigma' m''\vec{p}'''\vec{q}'''\sigma''} & \left(\frac{\langle c\vec{p}'+\vec{q}'\sigma'|e^{i\vec{q}''\cdot\vec{r}}\vec{p}\cdot\vec{e}_2|m'\vec{p}'\sigma'\rangle\langle m''\vec{p}''+\vec{q}''\sigma''|e^{i\vec{q}'''\cdot\vec{r}}\vec{p}\cdot\vec{e}_1|c\vec{p}''\sigma''\rangle}{E_c(\vec{p}) - \epsilon_n(\vec{k}) + \omega_1} \right. \\ & \left. + \frac{\langle m''\vec{p}''+\vec{q}''\sigma''|e^{i\vec{q}'''\cdot\vec{r}}\vec{p}\cdot\vec{e}_1|c\vec{p}''\sigma''\rangle\langle c\vec{p}'+\vec{q}'\sigma'|e^{i\vec{q}''\cdot\vec{r}}\vec{p}\cdot\vec{e}_2|m'\vec{p}'\sigma'\rangle}{E_c(\vec{p}) - \epsilon_n(\vec{k}) - \omega_2} \right) \\ & \times \langle 0_s|\gamma_{\vec{p}+\vec{q}\sigma}c^\dagger_{\vec{p}''+\vec{q}''\sigma'}c_{\vec{p}'''\sigma''}\gamma_{\vec{p}\sigma}|0_s\rangle\langle 0_n|c_{m'\vec{p}''\sigma'}c^\dagger_{n\vec{k}\delta}|0_n\rangle\langle 0_n|c_{n\vec{k}\delta}c^\dagger_{m''\vec{p}'''+\vec{q}'''\sigma''}|0_n\rangle \ . \end{aligned} \qquad (4.6)$$

By using Eqs. (2.2), (4.2), the anticommutation relations for γ's, c's, and summing over the possible spin configurations in Eq. (4.6) one finds only the following terms different from zero:

$$\begin{aligned} &\langle 0_n|c_{m'\vec{p}''\sigma'}c^\dagger_{n\vec{k}\delta}|0_n\rangle = \delta_{n,m'}\delta_{\vec{k},\vec{p}''}\delta_{\delta,\sigma'} \ , \\ &\langle 0_n|c_{n\vec{k}\delta}c^\dagger_{m''\vec{p}'''+\vec{q}'''\sigma''}|0_n\rangle = \delta_{n,m''}\delta_{\vec{k},\vec{p}'''+\vec{q}'''}\delta_{\delta,\sigma''} \ , \\ &\langle 0_s|\gamma_{\vec{p}+\vec{q}\uparrow}c^\dagger_{\vec{k}'\uparrow}c_{\vec{p}'''\uparrow}\gamma^\dagger_{\vec{p}\uparrow}|0_s\rangle = u_{\vec{p}}u_{\vec{p}+\vec{q}}\delta_{\vec{p}+\vec{q},\vec{k}'}\delta_{\vec{p},\vec{p}'''} \ , \\ &\langle 0_s|\gamma_{\vec{p}+\vec{q}\downarrow}c^\dagger_{\vec{k}'\uparrow}c_{\vec{p}'''\uparrow}\gamma^\dagger_{\vec{p}\downarrow}|0_s\rangle = v_{\vec{p}}v_{\vec{p}+\vec{q}}\delta_{\vec{p}+\vec{q},-\vec{p}'''}\delta_{\vec{p},-\vec{k}'} \ , \\ &\langle 0_s|\gamma_{\vec{p}+\vec{q}\uparrow}c^\dagger_{\vec{k}'\downarrow}c_{\vec{p}'''\downarrow}\gamma^\dagger_{\vec{p}\uparrow}|0_s\rangle = v_{\vec{p}}v_{\vec{p}+\vec{q}}\delta_{\vec{p}+\vec{q},-\vec{p}'''}\delta_{\vec{p},-\vec{k}'} \ , \\ &\langle 0_s|\gamma_{\vec{p}+\vec{q}\downarrow}c^\dagger_{\vec{k}'\downarrow}c_{\vec{p}'''\downarrow}\gamma^\dagger_{\vec{p}\downarrow}|0_s\rangle = u_{\vec{p}}u_{\vec{p}+\vec{q}}\delta_{\vec{p}+\vec{q},\vec{k}'}\delta_{\vec{p},\vec{p}'''} \ , \end{aligned} \qquad (4.7)$$

where $\vec{k}' \equiv \vec{p}' + \vec{q}'$.

At this stage we assume the following approximations are valid:

$$\begin{aligned} \langle c\vec{k}+\vec{q}\delta|e^{i\vec{q}\cdot\vec{r}}\vec{p}\cdot\vec{e}|n\vec{k}\delta\rangle &\simeq \langle c\vec{k}\delta|\vec{p}\cdot\vec{e}|n\vec{k}\delta\rangle \\ &\simeq \langle c\vec{k}-\delta|\vec{p}\cdot\vec{e}|n\vec{k}-\delta\rangle \\ &\equiv \langle c\vec{k}|\vec{p}\cdot\vec{e}|n\vec{k}\rangle \ . \end{aligned} \qquad (4.8)$$

Then with Eqs. (4.7), (4.8), (2.5), and after performing the summations in Eq. (4.6), one obtains the following expression for the transition matrix element

$$\langle f|t_{12}|i\rangle = M_N(\vec{p}, \omega_1, \omega_2)I(\vec{p}, \vec{q}) \ , \qquad (4.9)$$

where

$$M_N(\vec{p},\omega_1,\omega_2) \equiv \frac{2}{m}\sum_n \left[\frac{\langle c\vec{p}\,|\,\vec{p}\cdot\vec{e}_2\,|\,n\vec{p}\rangle\langle n\vec{p}\,|\,\vec{p}\cdot\vec{e}_1\,|\,c\vec{p}\rangle}{E_c(\vec{p})-\epsilon_n(\vec{p})+\omega_1} + \frac{\langle c\vec{p}\,|\,\vec{p}\cdot\vec{e}_1\,|\,n\vec{p}\rangle\langle n\vec{p}\,|\,\vec{p}\cdot\vec{e}_2\,|\,c\vec{p}\rangle}{E_c(\vec{p})-\epsilon_n(\vec{p})-\omega_2} \right] \tag{4.10}$$

and $l(\vec{p},\vec{q})$ is the usual coherence factor, given by

$$l(\vec{p},\vec{q}) = u_{\vec{p}}u_{\vec{p}+\vec{q}} + v_{\vec{p}}v_{\vec{p}+\vec{q}} = \frac{1}{\sqrt{2}}\left[1+\frac{\epsilon_c(\vec{p})\epsilon_c(\vec{p}+\vec{q})+\Delta^2}{E_c(\vec{p})E_c(\vec{p}+\vec{q})}\right]^{1/2} . \tag{4.11}$$

With Eqs. (4.9), (4.4), and (4.3) the scattering of light by one-electron excitations in a superconductor can be written in the form

$$\left[\frac{d^2\sigma}{d\Omega\, d\omega}\right]_N = r_0^2\frac{\omega_2}{\omega_1}\sum_{\vec{p}} |M_N(\vec{p},\omega_1,\omega_2)|^2 l^2(\vec{p},\vec{q}) \times [f(E_c(\vec{p})) - f(E_c(\vec{p}+\vec{q}))] \times \delta(\omega + E_c(\vec{p}) - E_c(\vec{p}+\vec{q})) \tag{4.12}$$

From Eq. (4.12) we see, that the scattering can occur only at $T \neq 0$ °K.

The processes of scattering are shown schematically in Fig. 2. a and c are the two configurations entering the wave function for a state with a quasiparticle in $\vec{p}\uparrow$. b and d are the two configurations which can be connected to a and c, respectively, by spin-independent one-body operator. The arrows indicate how $a \rightarrow b$ and $c \rightarrow d$ via intermediate interband states n, when electrons couple to the electromagnetic field which does not flip the electronic spin.

V. SCATTERING OF LIGHT BY TWO-ELECTRON EXCITATIONS

The "superfluid" scattering cross section involves creation of two quasiparticles with the minimum excitation energy being 2Δ.

The initial and final states and energies are given in this case by

$$|i|0_s\rangle = |0_n\rangle , \quad |f\rangle = \gamma^\dagger_{\vec{p}\sigma}\gamma^\dagger_{-\vec{p}-\vec{q}-\sigma}|0_s\rangle|0_n\rangle , \quad \sigma = \uparrow,\downarrow; \tag{5.1}$$

$$E_i = 0 , \quad E_f = E_c(\vec{p}) + E_c(\vec{p}+\vec{q}) . \tag{5.2}$$

The thermal weight rate factor becomes

$$P_i - P_f = [1-f(E_c(\vec{p}))][1-f(E_c(\vec{p}+\vec{q}))] - f(E_c(\vec{p}))f(E_c(\vec{p}+\vec{q})) = 1 - f(E_c(\vec{p})) - f(E_c(\vec{p}+\vec{q})) \tag{5.3}$$

and the transition matrix element assumes the form

$$\langle f|t_{12}|i\rangle = \frac{1}{m}\sum_{n\vec{k}\delta m'\vec{p}'\vec{q}'\sigma' m''\vec{p}''\sigma''} \left[\frac{\langle c\vec{p}'+\vec{q}'\sigma'|e^{i\vec{q}'\cdot\vec{r}}\vec{p}\cdot\vec{e}_2|m'\vec{p}'\sigma'\rangle\langle m''\vec{p}''+\vec{q}''\sigma''|e^{i\vec{q}''\cdot\vec{r}}\vec{p}\cdot\vec{e}_1|c\vec{p}''\sigma''\rangle}{0-\epsilon_n(\vec{k})+\omega_1} + \frac{\langle m''\vec{p}''+\vec{q}''\sigma''|e^{i\vec{q}''\cdot\vec{r}}\vec{p}\cdot\vec{e}_1|c\vec{p}''\sigma''\rangle\langle c\vec{p}'+\vec{q}'\sigma'|e^{i\vec{q}'\cdot\vec{r}}\vec{p}\cdot\vec{e}_2|m'\vec{p}'\sigma'\rangle}{0-\epsilon_n(\vec{k})-\omega_2} \right]$$
$$\times \langle 0_s|\gamma_{\vec{p}\sigma}\gamma_{-\vec{p}-\vec{q}-\sigma}c^\dagger_{\vec{p}'+\vec{q}'\sigma'}c_{\vec{p}''\sigma''}|0_s\rangle\langle 0_n|c_{m'\vec{p}'\sigma'}c^\dagger_{n\vec{k}\delta}|0_n\rangle\langle 0_n|c_{n\vec{k}\delta}c^\dagger_{m''\vec{p}''+\vec{q}''\sigma''}|0_n\rangle . \tag{5.4}$$

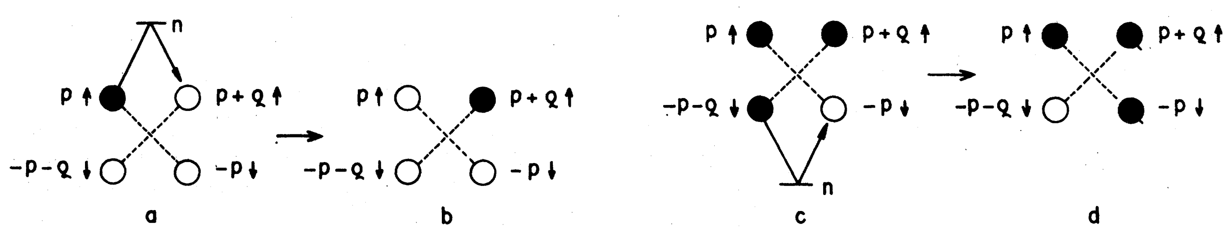

FIG. 2. Schematic representation of the inelastic scattering of light in a superconductor by one-electron excitations in the temperature interval $T_c > T > 0$ °K.

By using Eqs. (2.2), (4.2) the anticommutation relations for γ's, c's and summing over the possible spin configurations in Eqs. (5.4) one finds only the following terms different from zero

$$\langle 0_n | c_{m'\vec{p}'\sigma'} c^{\dagger}_{n\vec{k}\delta} | 0_n \rangle = \delta_{n,m'} \delta_{\vec{k},\vec{p}'} \delta_{\delta,\sigma'} ,$$

$$\langle 0_n | c_{n\vec{k}\delta} c^{\dagger}_{m''\vec{p}''+\vec{q}''\sigma''} | 0_n \rangle = \delta_{n,m''} \delta_{\vec{k},\vec{p}''+\vec{q}''} \delta_{\delta,\sigma''} ,$$

$$\langle 0_s | \gamma_{\vec{p}\uparrow} \gamma_{-\vec{p}-\vec{q}\downarrow} c^{\dagger}_{\vec{k}'\uparrow} c_{\vec{p}''\uparrow} | 0_s \rangle = u_{\vec{p}} v_{\vec{p}+\vec{q}} \delta_{\vec{p},\vec{k}'} \delta_{-\vec{p}-\vec{q},\vec{p}''} ,$$

$$\langle 0_s | \gamma_{\vec{p}\uparrow} \gamma_{-\vec{p}-\vec{q}\downarrow} c^{\dagger}_{-\vec{p}''\downarrow} c_{-\vec{k}'\downarrow} | 0_s \rangle = -v_{\vec{p}} u_{\vec{p}+\vec{q}} \delta_{\vec{p},\vec{p}''} \delta_{-\vec{p}-\vec{q},-\vec{p}'} ,$$

$$\langle 0_s | \gamma_{\vec{p}\downarrow} \gamma_{-\vec{p}-\vec{q}\uparrow} c^{\dagger}_{\vec{k}'\uparrow} c_{\vec{p}''\uparrow} | 0_s \rangle = v_{\vec{p}} u_{\vec{p}+\vec{q}} \delta_{\vec{p},-\vec{p}''} \delta_{-\vec{p}-\vec{q},\vec{k}'} ,$$

$$\langle 0_s | \gamma_{\vec{p}\downarrow} \gamma_{-\vec{p}-\vec{q}\uparrow} c^{\dagger}_{-\vec{k}'\downarrow} c_{-\vec{p}''\downarrow} | 0_s \rangle = -u_{\vec{p}} v_{\vec{p}+\vec{q}} \delta_{\vec{p},-\vec{k}'} \delta_{-\vec{p}-\vec{q},\vec{p}''} , \tag{5.5}$$

where $\vec{k}'' \equiv \vec{p}''$ and $\vec{k}' \equiv \vec{p}' + \vec{q}'$.

With Eqs. (5.5), (4.8), (4.2), and after performing the summation in Eq. (5.4), one obtains

$$\langle f | t_{12} | i \rangle = M_S(\vec{p}, \omega_1, \omega_2) p(\vec{p}, \vec{q}) , \tag{5.6}$$

where

$$M_S(\vec{p}, \omega_1, \omega_2) = \frac{2}{m} \sum_n \left(\frac{\langle c\vec{p} | \vec{p}\cdot\vec{e}_2 | n\vec{p} \rangle \langle n\vec{p} | \vec{p}\cdot\vec{e}_1 | c\vec{p} \rangle}{-\epsilon_n(\vec{p}) + \omega_1} + \frac{\langle c\vec{p} | \vec{p}\cdot\vec{e}_1 | n\vec{p} \rangle \langle n\vec{p} | \vec{p}\cdot\vec{e}_2 | c\vec{p} \rangle}{-\epsilon_n(\vec{p}) - \omega_2} \right) \tag{5.7}$$

and the coherence factor $p(\vec{p}, \vec{q})$ is given by

$$p(\vec{p}, \vec{q}) = u_{\vec{p}} v_{\vec{p}+\vec{q}} - v_{\vec{p}} u_{\vec{p}+\vec{q}} = \frac{1}{\sqrt{2}} \left(1 - \frac{\epsilon_c(\vec{p}) \epsilon_c(\vec{p}+\vec{q}) + \Delta^2}{E_c(\vec{p}) E_c(\vec{p}+\vec{q})} \right)^{1/2} . \tag{5.8}$$

With Eqs. (5.1), (5.2), (5.3), and (5.6), the scattering cross section for the inelastic scattering of light by two-electron excitations can be written as follows:

$$\left(\frac{d^2\sigma}{d\Omega\, d\omega} \right)_S = r_0^2 \frac{\omega_2}{\omega_1} \sum_{\vec{p}} | M_S(\vec{p}, \omega_1, \omega_2) |^2 p^2(\vec{p}, \vec{q}) \times [1 - f(E_c(\vec{p})) - f(E_c(\vec{p}+\vec{q}))] \times \delta(\omega - E_c(\vec{p}) - E_c(\vec{p}+\vec{q})) . \tag{5.9}$$

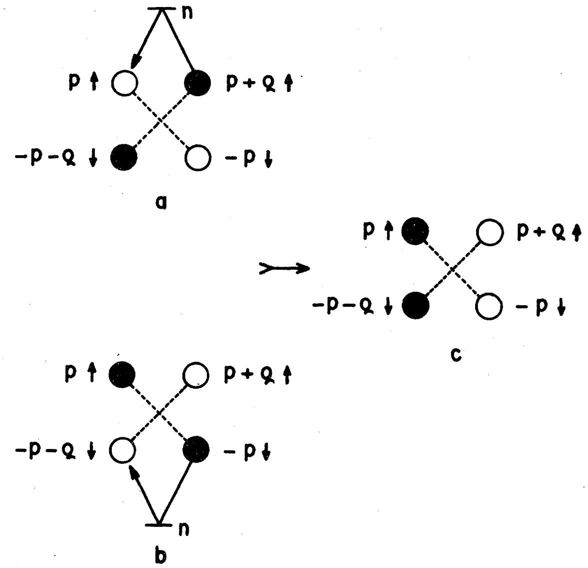

FIG. 3. Schematic representation of the inelastic scattering of light in a superconductor by two-electron excitations in the temperature interval $T_c > T \geq 0$ °K.

From Eq. (5.9) we see that the scattering is possible even at $T = 0$ °K.

Schematic representation for the scattering of light by two quasiparticle excitations is shown in Fig. 3. c is the only configuration entering the wave function for a state with quasiparticles in $\vec{p}\uparrow$ and $-\vec{p}-\vec{q}\downarrow$. a and b are the only two configurations which can be connected to that in a by a spin-independent one-body operator. The arrows indicate how $a \to c$ and $b \to c$ via intermediate interband states n, when electrons couple to the electromagnetic field which does not flip the spin.

VI. TOTAL SCATTERING CROSS SECTION

Clearly, the total scattering cross section is the sum

$$\left(\frac{d^2\sigma}{d\Omega\, d\omega} \right)_{\text{total}} = \left(\frac{d^2\sigma}{d\Omega\, d\omega} \right)_N + \left(\frac{d^2\sigma}{d\Omega\, d\omega} \right)_S . \tag{6.1}$$

From Eqs. (4.9) and (5.7) it is clear that the important intermediate states are the nonconducting states for which an energy denominator is small.

For conditions of large momentum transfer, i.e., $v_F | \vec{q} | \gg \omega_1 - \omega_2$ one can replace the integral over momenta in Eqs. (4.12) and (5.9) according to

$$\sum_{\vec{p}} \to \int \frac{d^3p}{(2\pi)^3} \to \frac{m^2}{(2\pi)^2 | \vec{q} |} \int dE_c(\vec{p}) \int dE_c(\vec{p}+\vec{q}) \frac{E_c(\vec{p}) [\omega + E_c(\vec{p})]}{[E_c^2(\vec{p}) - \Delta^2]^{1/2} \{[\omega + E_c(\vec{p})]^2 - \Delta^2\}^{1/2}} . \tag{6.2}$$

If the band structure and the incident frequency ω_1, are such that the matrix elements given by Eqs. (4.9) and (5.7) could be assumed constant, then the integration over one energy variable can be carried out easily, and one finds

$$\left[\frac{d^2\sigma}{d\Omega\,d\omega}\right]_N = r_0^2\frac{\omega_2}{\omega_1}\frac{m^2|M_N|^2}{(2\pi)^2}\int dE_c(\vec{p})\,[f(E_c(\vec{p}))-f(\omega+E_c(\vec{p}))]\frac{E_c(\vec{p})[\omega+E_c(\vec{p})]+\Delta^2}{[E_c^2(\vec{p})-\Delta^2]^{1/2}\{[\omega+E_c(\vec{p})]^2-\Delta^2\}^{1/2}} \tag{6.3}$$

and

$$\left[\frac{d^2\sigma}{d\Omega\,d\omega}\right]_S = r_0^2\frac{\omega_2}{\omega_1}\frac{m^2|M_S|^2}{(2\pi)^2}\int dE_c(\vec{p})\,[1-f(E_c(\vec{p}))-f(\omega-E_c(\vec{p}))]\frac{E_c(\vec{p})[\omega-E_c(\vec{p})]-\Delta^2}{[E_c^2(\vec{p})-\Delta^2]^{1/2}\{[\omega-E_c(\vec{p})]^2-\Delta^2\}^{1/2}} . \tag{6.4}$$

It is convenient to relate the scattering cross section of the superconductor to that in the normal metal at the same temperature. One can find the appropriate expression for the normal metal simply by putting $\Delta=0$ in Eq. (6.3).

For $M_N \simeq M_S \equiv M$ the relative total scattering cross section becomes

$$\frac{\tilde{\varphi}_s}{\tilde{\varphi}_n} \equiv \frac{\left[\frac{d^2\sigma}{d\Omega\,d\omega}\right]_N + \left[\frac{d^2\sigma}{d\Omega\,d\omega}\right]_S}{\left[\frac{d^2\sigma}{d\Omega\,d\omega}\right]_n} = \begin{cases} \dfrac{I_N+I_S}{I_n} & \text{for } \omega \geq 2\Delta \\ \dfrac{I_N}{I_n} & \text{for } \omega < 2\Delta \end{cases} , \tag{6.5}$$

where

$$I_N = \int_\Delta^\infty dE\,[f(E)-f(\omega+E)] \times \frac{E(\omega+E)+\Delta^2}{(E^2-\Delta^2)^{1/2}[(\omega+E)^2-\Delta^2]^{1/2}} , \tag{6.6}$$

$$I_S = \int_\Delta^{\omega-\Delta} dE\,[1-f(E)-f(\omega-E)] \times \frac{E[\omega-E]-\Delta^2}{(E^2-\Delta^2)^{1/2}[(\omega-E)^2-\Delta^2]^{1/2}} , \tag{6.7}$$

$$I_n = \int_0^\infty dE[f(E)-f(\omega+E)] . \tag{6.8}$$

For $T=0$ °K Eq. (6.5) can be written in terms of complete eliptic integrals as

$$\frac{\tilde{\varphi}_s}{\tilde{\varphi}_n} = \begin{cases} \dfrac{2\Delta+\omega}{\omega}E\left[\dfrac{2\Delta-\omega}{2\Delta+\omega}\right] - \dfrac{4\Delta}{\omega}K\left[\dfrac{2\Delta-\omega}{2\Delta+\omega}\right] \\ \quad \text{for } \omega \geq 2\Delta \\ 0 \quad \text{for } \omega < 2\Delta . \end{cases} \tag{6.9}$$

The results (6.5) and (6.9) are the same as obtained in Ref. 2 by a different method.

VIII. COMPARISON WITH THE EXPERIMENT

Because of the difficult estimation of the temperature of the spot where the focused laser beam strikes the sample the value of the gap was not determined from the usual gap equation but rather by the interpolation of the experimental results by Fraas *et al.* For the case of Nb_3Sn one gets $2\Delta = 14$ cm^{-1}.

The integrals in Eq. (6.5) were then solved numerically. Figure 4 shows a rather good agreement between the theoretical and experimental results. This indicates that the observed intensity of light in the frequency interval of interest comes from scattering rather than fluorescence.

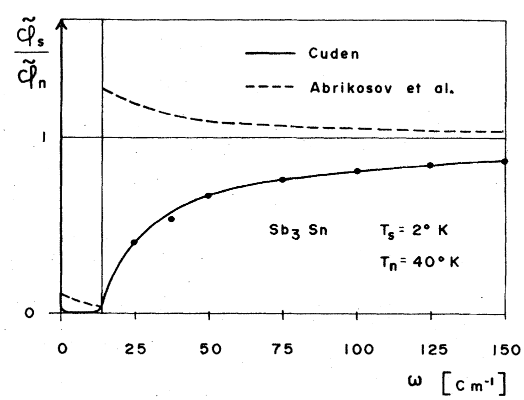

FIG. 4. Experimental data for the relative scattering efficiently in Sb_3Sn at $T_s = 2$ °K and $T_n = 40$ °K by Fraas *et al.* The solid and broken lines are the theoretical predictions by Cuden and Abrikosov *et al.*, respectively.

From Eqs. (4.9) and (5.7) one can see that depending on the band structure and the frequency of the incident light the denominators may vanish. In this case the virtual interband transitions may become real transitions. Then the electron in the nonconducting state could decay to the ground state via excited superconducting states. This process could give rise to the resonant fluorescence, and could presumably explain the appearance of the experimentally observed sharp peaks superimposed on the spectrum shown in Fig. 4.

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