Raman scattering of light off a superconducting alloy containing paramagnetic impurities

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Raman scattering of light off a superconductor doped with a small concentration of paramagnetic impurities is discussed without recourse to the quasiparticle approximation. The scattering efficiency defined in terms of Kubo's nonlinear response theory in a form suitable for systematic diagrammatic expansion is used as the starting point. This is examined in the four-component Eliashberg-Gorkov space for the case of constant transition-matrix elements, in the limit of small momentum transfer (London limit), and in the Eliashberg approximation of neglecting the momentum dependence of the electronic self-energy. For the specific case of Fe impurities in quenched In films numerical calculations have been made for the ratio of the scattering efficiency in the superconducting and normal states as a function of the reduced frequency and temperature for different impurity concentrations.

I. INTRODUCTION

In recent literature Raman scattering of light in pure superconductors was discussed. In Ref. 1 it was demonstrated by means of the standard formulation of Raman scattering of light that the main contribution to the intensity of the inelastically scattered light in a superconductor comes from the electronic interband intermediate transitions whose energy difference is close to the energy of the incident light. At finite temperatures such transitions are possible for the electrons in the normal and superfluid component.

In Ref. 2 the scattering efficiency was formulated using Kubo's nonlinear response theory in a form suitable for systematic diagrammatic expansion. The effects of the sample surface were taken into account by solving the proper boundaryvalue problem at the metal surface and introducing Fresnel correction factors. 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 considered in earlier literature.

In this work we calculate the scattering efficiency for the scattering of light off a superconducting alloy in the case of low concentration of randomly distributed paramagnetic impurities and weak conduction-electron-magnetic-impurity interaction. The physical situation when the orbital momentum of the impurity is quenched is considered only. In this case the electron scattering by the impurity is isotropic.

In Sec. II we give the system Hamiltonian. In Sec. III we represent the equilibrium one-particle Green's functions representing superconducting and nonconducting alloy states. In Sec. IV we discuss a general formalism for the inelastic scattering of light off a superconducting alloy surface and calculate the scattering efficiency in the limit of weak momentum transfer for various concentrations and temperatures. And finally, in Sec. V, we give our concluding remarks.

II. SYSTEM HAMILTONIAN

We take the Hamiltonian of the system to have the following form

$$H = \int d^{3}r \,\psi_{\alpha}^{\dagger}(\mathbf{\bar{r}})\epsilon_{c}(\mathbf{\bar{p}})\psi_{\alpha}(\mathbf{\bar{r}})$$

$$-\frac{g}{2} \int d^{3}r \,\psi_{\alpha}^{\dagger}(\mathbf{\bar{r}})\psi_{\beta}^{\dagger}(\mathbf{\bar{r}})\psi_{\beta}(\mathbf{\bar{r}})\psi_{\alpha}(\mathbf{\bar{r}})$$

$$+\sum_{j} \int d^{3}r \,\psi_{\alpha}^{\dagger}(\mathbf{\bar{r}})V_{\alpha\beta}(\mathbf{\bar{r}}-\mathbf{\bar{R}}_{j})\psi_{\beta}(\mathbf{\bar{r}})$$

$$+\sum_{n} \int d^{3}r \,\psi_{n\alpha}^{\dagger}(\mathbf{\bar{r}})\epsilon_{n}(\mathbf{\bar{p}})\psi_{n\alpha}(\mathbf{\bar{r}}), \qquad (1)$$

$$\mathbf{\bar{p}} = -i\nabla, \quad \epsilon_{c}(\mathbf{\bar{p}}) = \mathbf{\bar{p}}^{2}/2m_{0} - \mu.$$

The first term represents the kinetic energy of the electrons in a conduction band measured from the chemical potential μ . The second term represents the usual phonon-induced attraction between the electrons introduced by Gor'kov,³ the third term represents the interaction of an electron with a paramagnetic impurity of spin S at the position $\vec{\mathbf{R}}_{j}$, and the fourth term represents the energy of the electrons in the nonconducting bands. The short-ranged potential $V_{\alpha\beta}(\mathbf{\vec{r}})$ is taken to be of the form

$$V_{\alpha\beta}(\mathbf{\vec{r}}) = V_1(\mathbf{\vec{r}}) \delta_{\alpha\beta} + V_2(\mathbf{\vec{r}})(\mathbf{\vec{s}} \mathbf{\vec{\sigma}}_{\alpha\beta}/2), \qquad (2)$$

where $\vec{\sigma}$ is the Pauli spin matrix vector, and $V_1(\vec{r})$ and $V_2(\mathbf{r})$ are the strengths of the potential and exchange interaction, respectively.

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III. EQUILIBRIUM ONE-PARTICLE GREEN'S FUNCTIONS

We assume that the magnetic impurities are randomly distributed in a superconductor, and that their concentration is low enough so that impurity-impurity interaction can be neglected. The effects of the exchange interaction, which lead to large changes in order parameter and the attractive electron-electron interaction can be taken into account simultaneously by using four-component space introduced by Eliashberg.⁴ Namely, the Green's function 4×4 matrix characterizing the equilibrium properties of the superconducting alloy can be defined in this case as

$$\mathfrak{S}_{c}(x_{1}, x_{2}) = -i\langle T[\psi(x_{1})\psi^{\dagger}(x_{2})]\rangle$$
(3)

where the four-component annihilation and creation operators are given by

$$\psi(x) = \begin{pmatrix} \psi_{\dagger}(x) \\ \psi_{\downarrow}(x) \\ \psi_{\dagger}^{\dagger}(x) \\ \psi_{\dagger}^{\dagger}(x) \\ \psi_{\dagger}^{\dagger}(x) \end{pmatrix}, \qquad (4)$$
$$\psi^{\dagger}(x) = [\psi_{\dagger}^{\dagger}(x)\psi_{\dagger}^{\dagger}(x)\psi_{\dagger}(x)\psi_{\downarrow}(x)], \qquad (4)$$
$$x = \vec{\mathbf{r}}, t$$

and $\langle \rangle$ denotes the thermal average as well as the average over the possible impurity positions and spin configurations.

In the energy-momentum representation the renormalized single-particle Green's function 4×4 matrix of the superconducting alloy, $\mathcal{G}_c(\mathbf{\tilde{p}}, i\epsilon_l)$ averaged over the positions and the spin orientations of the impurities, can be written conveniently in the form

$$\begin{aligned} & \mathfrak{S}_{c}(\mathbf{\vec{p}}, i\epsilon_{1}) = i\tilde{\epsilon}_{1}(1\times1) - \epsilon_{c}(\mathbf{\vec{p}})(\tau_{3}\times1) + \tilde{\Delta}_{1}(\tau_{2}\times\sigma_{2}) \\ & \epsilon_{1} = (2l+1)\pi/\beta, \quad l = 0, \pm 1, \pm 2, \pm \cdots, \qquad (5) \\ & \beta \equiv 1/k_{B}T, \end{aligned}$$

where 1 is unit 2×2 matrix, τ_i and σ_i are Pauli matrices operating on the space composed of the electron and hole states and on the ordinary spin states, respectively, and $\epsilon_c(\mathbf{\vec{p}})$ is the single-particle energy measured from the chemical potential $\epsilon_c(\mathbf{\vec{p}}) = p^2/2 m_0 - \mu$.

In the first Born approximation, the renormalized frequency $\tilde{\epsilon}_i$ and order parameter $\tilde{\Delta}_i$ are given by the following set of coupled equations⁵

$$\tilde{\epsilon}_{l} = \epsilon_{l} + \frac{1}{2} \left(\frac{1}{\tau^{(1)}} + \frac{1}{\tau^{(2)}} \right) \frac{\tilde{\epsilon}_{l}}{(\tilde{\epsilon}_{l}^{2} + \tilde{\Delta}_{l}^{2})^{1/2}} , \qquad (6)$$

$$\tilde{\Delta}_{l} = \Delta + \frac{1}{2} \left(\frac{1}{\tau^{(1)}} - \frac{1}{\tau^{(2)}} \right) \frac{\tilde{\Delta}_{l}}{(\tilde{\epsilon}_{l}^{2} + \tilde{\Delta}_{l}^{2})^{1/2}} , \qquad (7)$$

where $\Delta(T, 0)$ is the temperature-dependent order parameter of pure superconductor, and $\tau^{(1)}$ and $\tau^{(2)}$ are the scattering lifetimes due to the direct potential and exchange scattering, respectively, and depend on the concentration of impurities n_i , the density of the normal single-particle states at the Fermi surface N(0), the spin of the impurity S, and on the interaction strengths V_1 and V_2 . Namely,

$$\frac{1}{\tau^{(1)}} = 2\pi n_i N(0) \int \frac{d\Omega}{4\pi} \left[|V_1(p_F, \theta)|^2 + \frac{1}{4} S(S+1)|V_2(p_F, \theta)|^2 \right], \quad (8)$$

$$= 2\pi n_i N(0) \int \frac{d\Omega}{4\pi} \left[|V_1(p_F, \theta)|^2 - \frac{1}{4} S(S+1)|V_2(p_F, \theta)|^2 \right].$$
(9)

Defining a new auxiliary parameter u_i by $u_i = \tilde{\epsilon}_i / \tilde{\Delta}_i$ Eqs. (6) and (7) reduce to the single equation

$$\frac{\epsilon_I}{\Delta} = u_I \left[1 - \zeta \frac{1}{(1 + u_I^2)^{1/2}} \right],$$
(10)

where

 τ (2)

$$\zeta = \frac{1}{\tau_s \Delta}, \quad \frac{1}{\tau_s} = \frac{1}{2} \left(\frac{1}{\tau^{(1)}} - \frac{1}{\tau^{(2)}} \right).$$
(11)

For further discussion we need to define at this stage also the single-particle Green's function matrix $S_n(\vec{p}, i\epsilon_i)$, representing nonconducting alloy states

$$\mathfrak{S}_{n}(\mathbf{\tilde{p}},i\epsilon_{l}) = \frac{1\times 1}{i\epsilon_{l} - \epsilon_{n}(\mathbf{\tilde{p}})}, \quad \epsilon_{l} = (2l+1)\pi/\beta,$$
$$l = 0, \pm 1, \pm 2, \pm \cdots . \quad (12)$$

IV. SCATTERING EFFICIENCY

We are primarily interested in the photon energy transfers in the range $0 < \omega \le 6\Delta$, i.e., in the region of low-lying electronic excitations where $\omega = \omega_0 - \omega_s \ll \omega_0, \omega_s$. The frequencies of the incident light ω_0 , scattered light ω_s , and the transferred frequency ω , are related to the corresponding wave vectors \vec{k}_0 , \vec{k} , and \vec{q} in the usual way,

$$\omega_{0} = c |\vec{\mathbf{k}}_{0}| = ck_{0}, \quad \omega_{s} = c |\vec{\mathbf{k}}| = ck,$$

$$\omega = c |\vec{\mathbf{q}}| = cq, \quad \vec{\mathbf{q}} = \vec{\mathbf{k}}_{0} - \vec{\mathbf{k}}.$$
(13)

Following the results and notation of Ref. 2 one can write the scattering efficiency of the superconducting alloy containing paramagnetic impurities in the form

$$\frac{1}{\varphi_{0}}\frac{d^{2}\varphi}{d\omega d\Omega} = \frac{4\omega_{s}\cos^{2}\theta}{S\omega_{0}c^{4}\cos\theta_{0}}\sum_{\substack{\alpha\beta\gamma\delta\\\alpha'\beta'\gamma'\delta'}}e_{\alpha}e_{\beta}T_{\alpha\alpha'}T^{*}_{\beta\beta'}G_{\alpha'\beta'\gamma'\delta'}(\vec{k},\vec{k}_{0})D_{\gamma'\gamma}(\vec{k}_{\parallel},ck)D^{*}_{\delta'\delta}(\vec{k}_{\parallel},ck)e_{\gamma}e_{\delta},$$
(14)

where

$$G_{\alpha\beta\gamma\delta}(\vec{\mathbf{k}},\vec{\mathbf{k}}_{0}) = \frac{1}{2\pi i} \frac{1}{1 - e^{-\beta\omega}} \sum_{j} \int \frac{dq_{z}}{2\pi} \frac{1}{|\gamma_{0} + \gamma_{s} - iq_{z}|^{2}} \lim_{\delta_{0} \to 0^{+}} \operatorname{Im} \Gamma^{j}_{\alpha\beta\gamma\delta}(k_{1z},k_{2z},q_{z};i\omega_{1},i\omega_{2},i\omega_{3}) \Big|_{\substack{i\omega_{1} \to \omega_{0} + i\delta_{0} \\ i\omega_{2} \to -\omega_{0} + i\delta_{0}}}$$
(15)

and

 $\mathrm{Im}\;\Gamma^{j}_{\alpha\beta\gamma\delta}(\bar{\mathbf{k}}_{1},\bar{\mathbf{k}}_{2},\bar{\mathbf{q}};i\omega_{1},i\omega_{2},i\omega_{3}) = \lim_{\epsilon\to 0^{+}} [\Gamma^{j}_{\alpha\beta\gamma\delta}(\bar{\mathbf{k}}_{1},\bar{\mathbf{k}}_{2},\bar{\mathbf{q}};i\omega_{1},i\omega_{2},i\omega_{3}=\omega+i\epsilon)$

$$-\Gamma_{\alpha\beta\gamma\delta}^{j}(\vec{k}_{1},\vec{k}_{2},\vec{q};i\omega_{1},i\omega_{2},i\omega_{3}=\omega-i\epsilon)].$$
(16)



FIG. 1. Diagramatic representation of the twelve topologically different averaged four-vertex parts representing the scattering of light by electronic excitations in a superconducting alloy containing paramagnetic impurities.

 $\Gamma_{\alpha\beta\gamma\delta}^{j}$ is the *j*th topologically different fourvertex part averaged over different possible configurations of paramagnetic impurities, and describe the scattering of light by one- and twoparticle excitations in a superconducting alloy. It is convenient to write the above averaged fourvertex parts in terms of averaged single-particle Green's functions and corresponding vertex corrections. In fact it can be shown that the major effect of the vertex corrections is to replace the inverse collision time by the correct transport value τ^{tr} .

In the lowest order, i.e., fourth order in electronic charge e, one can construct twelve topologically different averaged four-vertex parts shown in Fig. 1. External photon lines are attached to each four-vertex part for clarity of exposition. Double lines correspond to the Green's function matrix 9_c of a superconducting alloy given by Eqs. (5)-(7). Single lines denote the Green's function matrix 9_n representing intermediate nonconducting states. Open small circle denote partial vertex part γ connected to one external photon line only and is given in the long-wave-limit approximation by the expression

$$\gamma(\alpha; c, \mathbf{p}; n, \mathbf{p} + \mathbf{q}) = (e/m_0) \langle c\mathbf{p} \mid p_\alpha \mid n\mathbf{p} \rangle,$$

$$p_\alpha \equiv \mathbf{p} \cdot \mathbf{e}_\alpha$$
(17)

where $\langle c\bar{p} \mid p_{\alpha} \mid n\bar{p} \rangle$ is the matrix element of momentum between conducting and nonconducting states, and m_0 is the bare electron mass.

In the construction of each averaged four-vertex part shown in Fig. 1 the fact that the average of the product of Green's functions is not equal to the product of the averaged Green's functions is taken into account by the shaded partial square vertex part Φ . In the ladder approximation Φ satisfies the following integral equation

$$\begin{split} \Phi(\mathbf{\tilde{p}},i\epsilon_{l};\mathbf{\tilde{p}}+\mathbf{\tilde{q}},i\epsilon_{l}+i\omega_{3};\mathbf{\tilde{p}}'+\mathbf{\tilde{q}},i\epsilon_{l}'+i\omega_{3};\mathbf{\tilde{p}}',i\epsilon_{l}') \\ &= |V(\mathbf{\tilde{p}}-\mathbf{\tilde{p}}')|^{2}+\frac{1}{4\beta}\sum_{i\epsilon_{l}''}\int\frac{d^{3}p''}{(2\pi)^{3}}\mathrm{Tr}[|V(\mathbf{\tilde{p}}-\mathbf{\tilde{p}}'')|^{2} \\ &\times 9_{c}(\mathbf{\tilde{p}}''+\mathbf{\tilde{q}},i\epsilon_{l}''+i\omega_{3})\Phi(\mathbf{\tilde{p}}'',i\epsilon_{l}'';\mathbf{\tilde{p}}''+\mathbf{\tilde{q}},i\epsilon_{l}''+i\omega_{3};\mathbf{\tilde{p}}'+\mathbf{\tilde{q}},i\epsilon_{l}'+i\omega_{3};\mathbf{\tilde{p}}',i\epsilon_{l}')9_{c}(\mathbf{\tilde{p}}'',i\epsilon_{l}'')]. \end{split}$$

Diagrammatic representation of the above integral equation is shown in Fig. 2.

The analytical expression for the four-vertex parts corresponding to the first and seventh diagram of Fig. 1 are

$$\Gamma^{I}_{\alpha\beta\gamma\delta}(\vec{k}_{1},\vec{k}_{2},\vec{q};i\omega_{1},i\omega_{2},i\omega_{3}) = \frac{-1}{2\beta} \sum_{m,n\neq c} \int \frac{d^{3}b}{(2\pi)^{3}} M(m\alpha c,c\beta n,c\gamma m,n\delta c;\vec{p}) \\ \times \sum_{i\epsilon_{l}} \operatorname{Tr}[g_{c}(\vec{p},i\epsilon_{l})g_{n}(\vec{p}+\vec{k}_{1},i\epsilon_{l}+i\omega_{1})g_{c}(\vec{p}+\vec{q},i\epsilon_{l}+i\omega_{3})g_{m}(\vec{p}-\vec{k}_{2},i\epsilon_{l}-i\omega_{2})],$$
(19)

$$\Gamma_{\alpha\beta\gamma\delta}^{\mathrm{VII}}(\vec{\mathbf{k}}_{1},\vec{\mathbf{k}}_{2},\vec{\mathbf{q}};i\omega_{1},i\omega_{2},i\omega_{3}) = \frac{-1}{2\beta} \sum_{m,n\neq c} \int \frac{d^{3}p}{(2\pi)^{3}} \int \frac{d^{3}p'}{(2\pi)^{3}} M(m\,\alpha\,c,c\,\beta\,n,c\,\gamma\,m,n\delta c\,;\vec{\mathbf{p}},\vec{\mathbf{p}}') \\ \times \sum_{i\in_{l}i\in_{l}'} \mathrm{Tr}[g_{c}(\vec{\mathbf{p}},i\epsilon_{l})g_{n}(\vec{\mathbf{p}}+\vec{\mathbf{k}}_{1},i\epsilon_{l}+i\omega_{1})g_{c}(\vec{\mathbf{p}}+\vec{\mathbf{q}},i\epsilon_{l}+i\omega_{3}) \\ \times \Phi(\vec{\mathbf{p}},i\epsilon_{l};\vec{\mathbf{p}}+\vec{\mathbf{q}},i\epsilon_{l}+i\omega_{3};\vec{\mathbf{p}}'+\vec{\mathbf{q}},i\epsilon_{l}'+i\omega_{3};\vec{\mathbf{p}}',i\epsilon_{l}') \\ \times g_{c}(\vec{\mathbf{p}}'+\vec{\mathbf{q}},i\epsilon_{l}'+i\omega_{3})g_{m}(\vec{\mathbf{p}}'-\vec{\mathbf{k}}_{2},i\epsilon_{l}'-i\omega_{2})g_{c}(\vec{\mathbf{p}}',i\epsilon_{l}')],$$

(20)

(18)

 $M(m\alpha c, c\beta n, c\gamma m, n\delta c; \mathbf{p}, \mathbf{p}')$

$$= \left(\frac{e}{m_0}\right)^4 \langle m \vec{\mathbf{p}} | p_\alpha | c \vec{\mathbf{p}} \rangle \langle c \vec{\mathbf{p}}' | p_\beta | n \vec{\mathbf{p}}' \rangle$$
$$\times \langle c \vec{\mathbf{p}} | p_\gamma | m \vec{\mathbf{p}} \rangle \langle n \vec{\mathbf{p}}' | p_\delta | c \vec{\mathbf{p}}' \rangle .$$
(22)

An over-all factor of $\frac{1}{2}$ in Eqs. (19) and (20) and the factor $\frac{1}{4}$ in the second term of Eq. (18) come

 $M(m\alpha c, c\beta n, c\gamma m, n\delta c; \mathbf{\vec{p}})$

$$= \left(\frac{e}{m_0}\right)^4 \langle m\vec{p} \mid p_\alpha \mid c\vec{p} \rangle \langle c\vec{p} \mid p_\beta \mid n\vec{p} \rangle$$
$$\times \langle c\vec{p} \mid p_\gamma \mid m\vec{p} \rangle \langle n\vec{p} \mid p_\delta \mid c\vec{p} \rangle \quad (21)$$

and



from the redundancy of the four-component operators given by Eq. (4).

In what follows, however, we shall neglect the partial vertex corrections. The important contribution to the scattering efficiency will come then from the first six diagrams shown in Fig. 1 only. This will limit the reliability of our results to low impurity concentrations.

As a matter of convenience let us devide the spectral representation of the Green's function matrix $g_c(\mathbf{p}, i\epsilon_l)$ into two parts characterizing the positive and negative energy spectrum of a superconducting alloy in the following manner:

$$g_c(\vec{p}, i\epsilon_l) = g_c^+(\vec{p}, i\epsilon_l) + g_c^-(\vec{p}, i\epsilon_l), \qquad (23)$$



FIG. 2. Schematic representation of the integral equation for the shaded partial quark vertex part Φ .

where

$$\mathfrak{g}_c^+(\mathbf{\tilde{p}},i\epsilon_1) = \int \frac{d\omega}{2\pi} \frac{a_c(\mathbf{\tilde{p}},\omega)}{i\epsilon_1 - \omega}, \qquad (24)$$

$$\mathfrak{g}_{c}(\mathbf{\bar{p}},i\epsilon_{l})=-\int\frac{d\omega}{2\pi}\frac{a_{c}(\mathbf{\bar{p}},-\omega)}{i\epsilon_{l}+\omega},\qquad(25)$$

and the spectral density matrix $a_c(\vec{p},\omega)$ is defined by

$$a_{c}(\vec{\mathfrak{p}},\omega) = 2 \operatorname{Im} \mathfrak{g}_{c}(\vec{\mathfrak{p}},\omega-i0^{+})$$
$$= i \left[\mathfrak{g}_{c}(\vec{\mathfrak{p}},\omega+i0^{+}) - \mathfrak{g}_{c}(\vec{\mathfrak{p}},\omega-i0^{+}) \right]. \quad (26)$$

By the use of Eq. (23) the four-vertex part $\Gamma^{I}_{\alpha\beta\gamma\delta}$ given by Eq. (19) can be split into four parts as follows:

$$\Gamma_{\alpha\beta\gamma\delta}^{I}(\vec{k}_{1},\vec{k}_{2},\vec{q};i\omega_{1},i\omega_{2},i\omega_{3}) = \frac{-1}{2\beta} \sum_{m,n\neq c} \int \frac{d^{3}p}{(2\pi)^{3}} M(m\alpha c,c\beta n,c\gamma m,n\delta c;\vec{p}) \\ \times \sum_{i \in I} \operatorname{Tr} \left[g_{c}^{+}(\vec{p},i\epsilon_{1})g_{n}(\vec{p}+\vec{k}_{1},i\epsilon_{1}+i\omega_{1})g_{c}^{+}(\vec{p}+\vec{q},i\epsilon_{1}+i\omega_{3})g_{m}(\vec{p}-\vec{k}_{2},i\epsilon_{1}-i\omega_{2}) \right. \\ \left. + g_{c}^{-}(\vec{p},i\epsilon_{1})g_{n}(\vec{p}+\vec{k}_{1},i\epsilon_{1}+i\omega_{1})g_{c}^{+}(\vec{p}+\vec{q},i\epsilon_{1}+i\omega_{3})g_{m}(\vec{p}-\vec{k}_{2},i\epsilon_{1}-i\omega_{2}) \right. \\ \left. + g_{c}^{+}(\vec{p},i\epsilon_{1})g_{n}(\vec{p}+\vec{k}_{1},i\epsilon_{1}+i\omega_{1})g_{c}^{-}(\vec{p}+\vec{q},i\epsilon_{1}+i\omega_{3})g_{m}(\vec{p}-\vec{k}_{2},i\epsilon_{1}-i\omega_{2}) \right. \\ \left. + g_{c}^{-}(\vec{p},i\epsilon_{1})g_{n}(\vec{p}+\vec{k}_{1},i\epsilon_{1}+i\omega_{1})g_{c}^{-}(\vec{p}+\vec{q},i\epsilon_{1}+i\omega_{3})g_{m}(\vec{p}-\vec{k}_{2},i\epsilon_{1}-i\omega_{2}) \right. \\ \left. + g_{c}^{-}(\vec{p},i\epsilon_{1})g_{n}(\vec{p}+\vec{k}_{1},i\epsilon_{1}+i\omega_{1})g_{c}^{-}(\vec{p}+\vec{q},i\epsilon_{1}+i\omega_{3})g_{m}(\vec{p}-\vec{k}_{2},i\epsilon_{1}-i\omega_{2}) \right] \right.$$

$$\left. + g_{c}^{-}(\vec{p},i\epsilon_{1})g_{n}(\vec{p}+\vec{k}_{1},i\epsilon_{1}+i\omega_{1})g_{c}^{-}(\vec{p}+\vec{q},i\epsilon_{1}+i\omega_{3})g_{m}(\vec{p}-\vec{k}_{2},i\epsilon_{1}-i\omega_{2}) \right] \right.$$

The first two and the last two terms in Eq. (27) determine the Stokes and anti-Stokes part of the scattering efficiency, respectively. In what follows we shall be considering the Stokes part of the spectrum only. Then after combining Eqs. (12), (24), (25), and (27) one obtains the following expression for the fourvertex part

$$\Gamma^{I}_{\alpha\beta\gamma\delta}(\vec{\mathbf{k}}_{1},\vec{\mathbf{k}}_{2},\vec{\mathbf{q}};i\omega_{1},i\omega_{2},i\omega_{3})=\Gamma^{\prime\prime}_{\alpha\beta\gamma\delta}(\vec{\mathbf{k}}_{1},\vec{\mathbf{k}}_{2},\vec{\mathbf{q}};i\omega_{1},i\omega_{2},i\omega_{3})+\Gamma^{\prime}_{\alpha\beta\gamma\delta}(\vec{\mathbf{k}}_{1},\vec{\mathbf{k}}_{2},\vec{\mathbf{q}};i\omega_{1},i\omega_{2},i\omega_{3})$$

where

$$\Gamma_{\alpha\beta\gamma\delta}^{NI}(\vec{k}_{1},\vec{k}_{2},\vec{q};i\omega_{1},i\omega_{2},i\omega_{3}) = \frac{-1}{2\beta} \sum_{m,n\neq c} \int \frac{d^{3}p}{(2\pi)^{3}} M(m\alpha c,c\beta n,c\gamma m,n\delta c;\vec{p}) \\ \times \int \frac{d\epsilon_{1}}{2\pi} \int \frac{d\epsilon_{2}}{2\pi} \operatorname{Tr}\left[a_{c}(\vec{p},\epsilon_{1})(1\times1)a_{c}(\vec{p}+\vec{q},\epsilon_{2})(1\times1)\right] \\ \times \sum_{i\epsilon_{l}} \frac{1}{i\epsilon_{l}-\epsilon_{1}} \frac{1}{i\epsilon_{l}+i\omega_{1}-\epsilon_{n}(\vec{p}+\vec{k}_{1})} \\ \times \frac{1}{i\epsilon_{l}+i\omega_{3}-\epsilon_{2}} \frac{1}{i\epsilon_{l}-i\omega_{2}-\epsilon_{m}(\vec{p}-\vec{k}_{2})},$$
(29)

(28)

$$\Gamma_{\alpha\beta\gamma\delta}^{S1}(\vec{k}_{1},\vec{k}_{2},\vec{q};i\omega_{1},i\omega_{2},i\omega_{3}) = \frac{1}{2\beta} \sum_{m,n\neq c} \int \frac{d^{3}p}{(2\pi)^{3}} M(m\,\alpha\,c,c\,\beta\,n,c\,\gamma\,m,n\delta c\,;\vec{p}) \\ \times \int \frac{d\epsilon_{1}}{2\pi} \int \frac{d\epsilon_{2}}{2\pi} \operatorname{Tr}\left[a_{c}(\vec{p},-\epsilon_{1})(1\times1)a_{c}(\vec{p}+\vec{q},\epsilon_{2})(1\times1)\right] \\ \times \sum_{i\epsilon_{l}} \frac{1}{i\epsilon_{l}+\epsilon_{1}} \frac{1}{i\epsilon_{l}+\epsilon_{1}} \frac{1}{i\epsilon_{l}+i\omega_{1}-\epsilon_{n}(\vec{p}+\vec{k}_{1})} \\ \times \frac{1}{i\epsilon_{l}+i\omega_{3}-\epsilon_{2}} \frac{1}{i\epsilon_{l}-i\omega_{2}-\epsilon_{m}(\vec{p}-\vec{k}_{2})}.$$
(30)

The frequency sums in Eqs. (29) and (30) can now be easily performed, and one obtains

$$\Gamma_{\alpha\beta\gamma\delta}^{NI}(\vec{k}_{1},\vec{k}_{2},\vec{q};i\omega_{1},i\omega_{2},i\omega_{3}) = \frac{1}{4} \int \frac{d^{3}p}{(2\pi)^{3}} |\mathfrak{M}_{\alpha\beta\gamma\delta}^{NI}(\vec{p},\omega_{0})|^{2} \int \frac{d\epsilon_{1}}{2\pi} \int \frac{d\epsilon_{2}}{2\pi} \left(\tanh\frac{\beta\epsilon_{2}}{2} - \tanh\frac{\beta\epsilon_{1}}{2} \right) \frac{1}{\omega_{3} + \epsilon_{1} - \epsilon_{2}} \times \operatorname{Tr}\left[a_{c}(\vec{p},\epsilon_{1})(1\times1)a_{c}(\vec{p}+\vec{q},\epsilon_{2})(1\times1) \right], \quad (31)$$

and

$$\Gamma_{\alpha\beta\gamma\delta}^{SI}(\vec{k}_{1},\vec{k}_{2},\vec{q};i\omega_{1},i\omega_{2},i\omega_{3}) = -\frac{1}{4} \int \frac{d^{3}p}{(2\pi)^{3}} |\mathfrak{M}_{\alpha\beta\gamma\delta}^{SI}(\vec{p},\omega_{0})|^{2} \int \frac{d\epsilon_{1}}{2\pi} \int \frac{d\epsilon_{2}}{2\pi} \left(\tanh\frac{\beta\epsilon_{2}}{2} + \tanh\frac{\beta\epsilon_{1}}{2} \right) \frac{1}{\omega_{3} - \epsilon_{1} - \epsilon_{2}} \times \operatorname{Tr}\left[a_{c}(\vec{p},-\epsilon_{1})(1\times1)a_{c}(\vec{p}+\vec{q},\epsilon_{2})(1\times1) \right], \quad (32)$$

where $\mathfrak{M}^{\textit{NI}}_{\alpha\beta\gamma\delta}$ and $\mathfrak{M}^{\textit{SI}}_{\alpha\beta\gamma\delta}$ are defined in the following manner:

$$\left|\mathfrak{M}_{\alpha\beta\gamma\delta}^{NI}(\vec{p},\omega_{0})\right|^{2} \equiv \lim_{\delta_{0}\to0^{+}} \sum_{m,n\neq c} \frac{M(m\alpha c, c\beta n, c\gamma m, n\delta c; \vec{p})}{[i\omega_{1}+\tilde{E}_{c}(\vec{p})-\epsilon_{n}(\vec{p})][-i\omega_{2}+\tilde{E}_{c}(\vec{p})-\epsilon_{m}(\vec{p})]} \left|_{\substack{i\omega_{1}\to\omega_{0}+i\delta_{0}\\i\omega_{2}\to\omega_{0}+i\delta_{0}}}\right|$$
(33)

$$|\mathfrak{M}_{\alpha\beta\gamma\delta}^{s_{\mathrm{I}}}(\vec{p},\omega_{0})|^{2} \equiv \lim_{\delta \to 0^{+}} \sum_{m,n\neq c} \frac{M(m\alpha c, c\beta n, c\gamma m, n\delta c; \vec{p})}{[i\omega_{1} - \tilde{E}_{c}(\vec{p}) - \epsilon_{n}(\vec{p})][-i\omega_{2} - \tilde{E}_{c}(\vec{p}) - \epsilon_{m}(\vec{p})]} \bigg|_{\substack{i\omega_{1} \to \omega_{0} + i\delta_{0} \\ i\omega_{2} \to -\omega_{0} + i\delta_{0}}}$$

and

$$\tilde{E}_c(\vec{\mathbf{p}}) = [\epsilon_c^2(\vec{\mathbf{p}}) + \tilde{\Delta}^2(\vec{\mathbf{p}})]^{1/2}.$$

In Eqs. (33) and (34) the dependence of the energy denominators on \vec{k}_1 , \vec{k}_2 , and \vec{q} is neglected. As the important intermediate states are far from the Fermi surface, the contributions to the frequency sum from the poles of the nonconducting Green's functions in Eqs. (31) and (32) were neglected as well. The imaginary parts of $\Gamma_{\alpha\beta\gamma\delta}^{NI}$ and $\Gamma_{\alpha\beta\gamma\delta}^{SI}$ can now be easily found to be of the form

$$\operatorname{Im} \Gamma_{\alpha\beta\gamma\delta}^{NI}(\vec{k}_{1},\vec{k}_{2},\vec{q};i\omega_{1},i\omega_{2},i\omega_{3}) = \frac{1}{4} \int \frac{d^{3}p}{(2\pi)^{3}} \int \frac{d\epsilon}{2\pi} |\mathfrak{M}_{\alpha\beta\gamma\delta}^{NI}(\vec{p},\omega_{0})|^{2} \left(\tanh \frac{\beta(\epsilon+\omega)}{2} - \tanh \frac{\beta\epsilon}{2} \right) \\ \times \operatorname{Tr} \left[a_{c}(\vec{p},\epsilon)(1\times1)a_{c}(\vec{p}+\vec{q},\epsilon+\omega)(1\times1) \right],$$
(36)
$$\operatorname{Im} \Gamma_{\alpha\beta\gamma\delta}^{SI}(\vec{k}_{1},\vec{k}_{2},\vec{q};i\omega_{1},i\omega_{2},i\omega_{3}) = -\frac{1}{4} \int \frac{d^{3}p}{(2\pi)^{3}} \int \frac{d\epsilon}{2\pi} |\mathfrak{M}_{\alpha\beta\gamma\delta}^{SI}(\vec{p},\omega_{0})|^{2} \left(\tanh \frac{\beta(\epsilon+\omega)}{2} + \tanh \frac{\beta\epsilon}{2} \right) \\ \times \operatorname{Tr} \left[a_{c}(\vec{p},-\epsilon)(1\times1)a_{c}(\vec{p}+\vec{q},\epsilon+\omega)(1\times1) \right].$$
(37)

By using Eqs. (5), (10), (16), and (26) and performing the trace in Eqs. (36) and (37) leads to the following form for the four-vertex part:

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(35)

$$\begin{split} \operatorname{Im} \Gamma_{\alpha \beta \gamma \delta}^{\mathrm{I}}(\vec{k}_{1}, \vec{k}_{2}, \vec{q}; i\omega_{1}, i\omega_{2}, i\omega_{3}) &= \int \frac{d^{3}p}{(2\pi)^{3}} \int_{a}^{b} d\epsilon | \mathfrak{M}_{\alpha \beta \gamma \delta}^{N\mathrm{I}}(\vec{p}, \omega_{0})|^{2} \left(\tanh \frac{\beta(\epsilon + \omega)}{2} - \tanh \frac{\beta \epsilon}{2} \right) \\ & \times \left[\operatorname{Im} \left(\frac{u}{(1 - u^{2})^{1/2}} \right) \operatorname{Im} \left(\frac{u'}{(1 - u'^{2})^{1/2}} \right) + \operatorname{Im} \left(\frac{1}{(1 - u^{2})^{1/2}} \right) \operatorname{Im} \left(\frac{1}{(1 - u'^{2})^{1/2}} \right) \right], \end{split}$$

where $u = u(\mathbf{p}, \epsilon)$ and $u' = u'(\mathbf{p} + \mathbf{q}, \epsilon + \omega)$.

In the above expressions ω is related to real frequencies by

$$\frac{\omega}{\Delta} = u \left(1 - \zeta \frac{1}{(1 - u^2)^{1/2}} \right)$$
(40)

and ω_s is defined as the maximum value of ω in the region 0 < u < 1, i.e.,

$$\frac{1}{\Delta} \frac{\partial \omega}{\partial u} = 1 - \zeta (1 - u^2)^{-3/2} = 0$$
(41)

which leads to

$$\omega_{g} = \begin{cases} \Delta (1 - \zeta^{2/3})^{3/2} & \text{for } \zeta < 1, \\ 0 & \text{for } \zeta > 1. \end{cases}$$
(42)

The two limiting cases of interest can now be distinguished: (a) Pippard limit or extreme anomalous skin effect region of large momentum transfer. In this case $1/\tau_s \ll v_F q$, and one can neglect $1/\tau_s$. Then the effect of the impurities dissappears

and one can treat the superconductor as a pure metal. Obviously one then gets the results of Cuden.² (b) London limit, or the normal skin effect region of small momentum transfer where $1/\tau_s \gg v_F q$. One can then neglect q dependence in Eqs. (38) and (39). This is the case we are interested in.

The remaining integral over the energy and momenta, Eqs. (38) and (39), is still rather intractable. It requires the detailed knowledge of the band structure. For simplicity, we shall assume that the frequency of the incident and reflected light, and the band structure are such that no resonant scattering can occur. In the frequency interval of interest, the matrix elements $\mathfrak{M}_{\alpha\beta\gamma\delta}^{NI}$ and $\mathfrak{M}_{\alpha\beta\gamma\delta}^{SI}$ are then slowly varying functions of the energy and momenta, and can be assumed constant. Further, if we make the weak momentum approximation introduced by Eliashberg, that is, if we neglect the momentum dependence of u and u', the integration over momenta can be easily carried out and one obtains

$$\operatorname{Im} \Gamma_{\alpha \beta \gamma \delta}^{\mathrm{I}}(\bar{\mathbf{k}}_{1}, \bar{\mathbf{k}}_{2}, \bar{\mathbf{q}}; i\omega_{1}, i\omega_{2}, i\omega_{3}) = |\mathfrak{M}^{N\mathrm{I}}|^{2} N(0) \int d\epsilon \left(\tanh \frac{\beta(\epsilon + \omega)}{2} - \tanh \frac{\beta \epsilon}{2} \right) \\ \times \left[\operatorname{Im} \left(\frac{u}{(1 - u^{2})^{1/2}} \right) \operatorname{Im} \left(\frac{1}{(1 - u^{2})^{1/2}} \right) + \operatorname{Im} \left(\frac{1}{(1 - u^{2})^{1/2}} \right) \operatorname{Im} \left(\frac{1}{(1 - u^{2})^{1/2}} \right) \right] \\ + |\mathfrak{M}^{S\mathrm{I}}|^{2} N(0) \int d\epsilon \left(\tanh \frac{\beta(\epsilon + \omega)}{2} + \tanh \frac{\beta \epsilon}{2} \right) \\ \times \left[\operatorname{Im} \left(\frac{u}{(1 - u^{2})^{1/2}} \right) \operatorname{Im} \left(\frac{u'}{(1 - u'^{2})^{1/2}} \right) - \operatorname{Im} \left(\frac{1}{(1 - u^{2})^{1/2}} \right) \operatorname{Im} \left(\frac{1}{(1 - u'^{2})^{1/2}} \right) \right] \\ \operatorname{for} \omega > \omega_{n} \quad (43)$$

for $\omega < \omega_{g}$ (39)

$$Im \Gamma_{\alpha \beta \gamma \delta}^{I}(\vec{k}_{1}, \vec{k}_{2}, \vec{q}; i\omega_{1}, i\omega_{2}, i\omega_{3}) = |\mathfrak{M}^{NI}|^{2} N(0) \int d\epsilon \left(\tanh \frac{\beta(\epsilon + \omega)}{2} - \tanh \frac{\beta \epsilon}{2} \right) \\ \times \left[Im \left(\frac{u}{(1 - u^{2})^{1/2}} \right) Im \left(\frac{u'}{(1 - u'^{2})^{1/2}} \right) + Im \left(\frac{1}{(1 - u^{2})^{1/2}} \right) Im \left(\frac{1}{(1 - u'^{2})^{1/2}} \right) \right] \\ for \ \omega < \omega_{g}$$
(44)

where $u = u(p_F, \epsilon)$ and $u' = u'(p_F, \epsilon + \omega)$.

Similar calculations can be done for the remaining five square vertex parts. Within the approximations that led to Eqs. (43) and (44) one can write

$$\operatorname{Im} \Gamma_{\alpha \beta \gamma \delta}(\vec{k}_{1}, \vec{k}_{2}, \vec{q}; i\omega_{1}, i\omega_{2}, i\omega_{3})$$

$$= \sum_{j=1}^{VI} \left[\operatorname{Im} \Gamma_{\alpha \beta \gamma \delta}^{Nj}(\vec{k}_{1}, \vec{k}_{2}, \vec{q}; i\omega_{1}, i\omega_{2}, i\omega_{3}) + \operatorname{Im} \Gamma_{\alpha \beta \gamma \delta}^{Sj}(\vec{k}_{1}, \vec{k}_{2}, \vec{q}; i\omega_{1}, i\omega_{2}, i\omega_{3}) \right]$$

$$(45)$$

and for

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$$\sum_{j=1}^{\text{VI}} |\mathfrak{M}^{Nj}|^2 \simeq \sum_{j=1}^{\text{VI}} |\mathfrak{M}^{Sj}|^2$$
(46)

the relative scattering efficiency can be expressed in the following form

$$\frac{\tilde{\varphi}_{s}}{\tilde{\varphi}_{n}} = \frac{(d^{2}\varphi/d\omega d\Omega)_{s} + (d^{2}\varphi/d\omega d\Omega)_{N}}{(d^{2}\varphi/d\omega d\Omega)_{n}} \\
= \begin{cases} (I_{N} + I_{s})/I_{n} & \text{for } \omega > \omega_{g} \\ I_{N}/I_{n} & \text{for } \omega < \omega_{g} , \end{cases}$$
(47)

where

$$\begin{split} I_{N} &= \int_{a}^{b} d\epsilon \left(\tanh \frac{\beta(\epsilon + \omega)}{2} - \tanh \frac{\beta \epsilon}{2} \right) \\ &\times \left[\operatorname{Im} \left(\frac{u}{(1 - u^{2})^{1/2}} \right) \operatorname{Im} \left(\frac{u'}{(1 - u'^{2})^{1/2}} \right) \\ &+ \operatorname{Im} \left(\frac{1}{(1 - u^{2})^{1/2}} \right) \operatorname{Im} \left(\frac{1}{(1 - u'^{2})^{1/2}} \right) \right], \end{split}$$
(48)

$$I_{S} = \int_{c}^{d} d\epsilon \left(\tanh \frac{\beta(\epsilon + \omega)}{2} + \tanh \frac{\beta\epsilon}{2} \right) \\ \times \left[\operatorname{Im} \left(\frac{u}{(1 - u^{2})^{1/2}} \right) \operatorname{Im} \left(\frac{u'}{(1 - u'^{2})^{1/2}} \right) \\ - \operatorname{Im} \left(\frac{1}{(1 - u^{2})^{1/2}} \right) \operatorname{Im} \left(\frac{1}{(1 - u'^{2})^{1/2}} \right) \right] ,$$
(49)

$$I_n = \int_0^\infty d\epsilon \left(\tanh \frac{\beta(\epsilon + \omega)}{2} - \tanh \frac{\beta\epsilon}{2} \right).$$
 (50)

Depending on the parameter ζ the integration limits in Eqs. (48) and (49) assume the following values:

$$(a,b) = \begin{cases} (\omega_g, \infty) & \text{for } \zeta < 1, \\ (0, \infty) & \text{for } \zeta \ge 1; \end{cases}$$
(51)

$$(c,d) = \begin{cases} (\omega_{\varepsilon}, \omega - \omega_{\varepsilon}) & \text{for } \zeta < 1, \\ (0,\omega) & \text{for } \zeta \ge 1. \end{cases}$$
(52)

Making use of the expression for u, one can distinguish the following different cases for ω close to the threshold

$$\operatorname{Im}\left(\frac{1}{(1-u^{2})^{1/2}}\right) = \begin{cases} \left(\frac{2}{3}\right)^{1/2} \zeta^{-2/3} (1-\zeta^{2/3})^{1/4} [(\omega-\omega_{g})/\Delta]^{1/2} \\ \text{for } \zeta < 1, \omega \ge \omega_{g}, \omega \simeq \omega_{g} \\ (\sqrt{3}/4) [(2\omega/\Delta)^{2/3} - \frac{1}{4} (2\omega/\Delta)^{4/3}] \\ \text{for } \zeta = 1, \omega \ge 0 \\ \zeta^{-2} (1-\zeta^{-2}) \omega/\Delta \\ \text{for } \zeta > 1, \omega \ge 0 \end{cases}$$
(53)

and

$$\operatorname{Im}\left(\frac{u}{(1-u^{2})^{1/2}}\right) = \begin{cases} \left(\frac{2}{3}\right)^{1/2} \zeta^{-2/3} (1-\zeta^{2/3})^{1/4} [(\omega-\omega_{g})/\Delta]^{1/2} \\ \text{for } \zeta < 1, \, \omega \ge \omega_{g}, \, \omega \simeq \omega_{g} \\ (\sqrt{3}/2) [(2\omega/\Delta)^{1/3} - \frac{1}{24} (2\omega/\Delta)^{5/3}] \\ \text{for } \zeta = 1, \, \omega \ge 0 \\ (1-\zeta^{-2})^{1/2} + \frac{3}{2} \zeta^{-4} (1-\zeta^{-2})^{-5/2} (\omega/\Delta)^{2} \\ \text{for } \zeta \ge 1, \, \omega \ge 0 . \end{cases}$$

Finally, in the extreme gapless region where Δ is a small parameter u is very large, and one can solve Eq. (40) expanding in inverse powers of u and obtain

$$\operatorname{Im}\left(\frac{1}{(1-u^2)^{1/2}}\right) = \frac{1}{\zeta} \frac{\omega}{\Delta} \frac{\operatorname{Im}u}{(\operatorname{Re}u)^2 + (\operatorname{Im}u)^2}$$
for $\zeta > 1, \omega \ge 0$, (55)

where

$$\operatorname{Re} u = \frac{\omega}{\Delta} + \frac{1}{2} \frac{\zeta^2 \Delta^2 \omega}{(\omega^2 + \zeta^2 \Delta^2)^2} , \qquad (56)$$

$$\operatorname{Im} u = \zeta + \frac{\zeta \Delta \omega^2 - \zeta^3 \Delta^3}{2(\omega^2 + \zeta^2 \Delta^2)^2}, \qquad (57)$$

and





$$\operatorname{Im}\left(\frac{u}{(1-u^{2})^{1/2}}\right) = 1 + \frac{\Delta^{2}}{2} \frac{\omega^{2} - \zeta^{2} \Delta^{2}}{(\omega^{2} + \zeta^{2} \Delta^{2})^{2}} + \frac{\Delta^{4}}{2} \left[\frac{3}{4} \frac{\omega^{4} - 6\zeta^{2} \Delta^{2} \omega^{2} + \zeta^{4} \Delta^{4}}{(\omega^{2} + \zeta^{2} \Delta^{2})^{4}} - \zeta^{2} \Delta^{2} \frac{5\omega^{4} - 10\zeta^{2} \Delta^{2} \omega^{2} + \zeta^{4} \Delta^{4}}{(\omega^{2} + \zeta^{2} \Delta^{2})^{5}}\right]$$
for $\zeta > 1, \omega \ge 0.$ (58)

Order parameter $\Delta(T, \zeta)$ for a paramagnetic alloy (relative to its value in a pure metal at T = 0 °K) as a function of the reduced temperature (relative to the critical temperature of a pure material T_{cp}) was given by Ambegaokar and Griffin.⁶ For low temperatures they found

$$\Delta(T,\zeta) = \begin{cases} \Delta(0,\zeta) - (\frac{2}{3}\pi)^{1/2} (\omega_g^{1/6}/\zeta^{2/3}\Delta^{2/3}) (k_BT)^{3/2} (1 - \frac{1}{4}\pi\zeta)^{-1} e^{-\omega_g/k_BT} & \text{for } \zeta < 1 \\ \Delta(0,\zeta) - (2^{2/3}/3^{1/2}) [(k_BT)^{5/3}/\Delta^{2/3}] (1 - \frac{1}{4}\pi)^{-1} \Gamma(\frac{2}{3}) \zeta(\frac{5}{3}) (1 - 2^{-2/3}) & \text{for } \zeta = 1 \\ \Delta(0,\zeta) - \frac{1}{6}\pi^2 (1 - \zeta^{-2})^{1/2} [1 - \frac{1}{2}(1 - \zeta^{-2})^{1/2} - \frac{1}{2}\zeta \arcsin\zeta^{-1}]^{-1} (k_BT)^2 / \Delta & \text{for } \zeta > 1, \zeta \simeq 1 \end{cases}$$
(59)

where $\Delta(0, \zeta)$ is the order parameter at T = 0 °K of a superconducting alloy determined by the following equation⁵:

$$\ln\left[\frac{\Delta(0,\zeta)}{\Delta(0,0)}\right] = \begin{cases} -\frac{1}{4}\pi\zeta & \text{for } \zeta \leq 1\\ -\arccos\zeta - \frac{1}{2}[\zeta \arcsin\zeta^{-1} - (1-\zeta^{-2})^{1/2}] & \text{for } \zeta > 1. \end{cases}$$
(60)

On the other hand for temperatures close to the critical temperature the concentration and temperaturedependent order parameter is given by the expression^{6,7}

$$\ln\frac{T_{c\rho}}{T} = \chi(\rho_s) + x_0^{-2} \sum_{l=0}^{\infty} \left(\frac{1}{(2l+1+\rho_s)} - \frac{\rho_s}{(2l+1+\rho_s)^4} \right) - \frac{3}{4} x_0^{-4} \sum_{l=0}^{\infty} \left(\frac{1}{(2l+1+\rho_s)^5} - \frac{3\rho_s}{(2l+1+\rho_s)^6} + \frac{2\rho_s}{(2l+1+\rho_s)^7} \right), \quad (61)$$

where

$$\psi(x) = \psi\left(\frac{1+x}{2}\right) - \psi(\frac{1}{2}), \quad \rho_s \equiv (\pi k_B T \tau_s)^{-1}$$

with $\psi(x)$ being the well known digamma function.

Equation (61) takes the simple form in the highand low-concentration limit, namely,

$$\Delta^{2}(T,\zeta) = \frac{8\pi^{2}k_{B}^{2}T^{2}}{7\zeta(3)} \left(\frac{T_{c}-T}{T_{c}}\right)$$

for $\rho_{Sc} \equiv (\pi k_{B}T_{c}\tau_{s})^{-1} \ll 1$ (62)

and

$$\Delta^{2}(T,\zeta) = 2\pi^{2} [(k_{B}T_{c})^{2} - (k_{B}T)^{2}] \text{ for } \rho_{sc} \gg 1.$$
 (63)

Combining the results for the order parameter $\Delta(T, \zeta)$ of Refs. 5 and 6 with Eqs. (47)–(58) the frequency dependence of the relative scattering efficiency was obtained numerically for various values of temperatures and concentrations of Fe impurities in quenched In films. In numerical calculations the BCS expression for the order parameter $\Delta(0,0)$ was used, namely, $\Delta(0,0) = 1.76k_BT_{cp}$ where T_{cp} is the transition temperature for In at $\eta_i = 0$. The results are plotted in Figs. 3(a), 3(b), and 3(c).

V. DISCUSSION

It is evident from our results that for temperatures and concentrations low enough there exists a sharp threshold in the spectrum of the scattered light at $2\omega_{x^*}$. In fact, this is the consequence of treating the self-energy resulting from the electron-impurity interaction in the first Born approximation.⁵ By means of the inelastic scattering of light one can thus check directly the well-known fact that with increasing concentration of paramagnetic impurities the energy gap is decreased relative to the energy gap of pure superconductors and may even vanish at high enough concentration of impurities.^{5,8}

It would be interesting to compare the energy gap obtained from the scattering experiments with the gap deduced from the tunneling measurements for Fe-In alloys by Reif and Woolf.⁸

Measurements of the transition temperature as a function of impurity concentration, for impurity concentration close to the critical concentration indicate systematic deviation from the theory.⁹⁻¹¹ The origin of this discrepancy is not clear at present although it may be partially due to the onset of a ferromagnetic or antiferromagnetic order among the impurity spins.

In this work we do not consider the effect of the indirect impurity spin interaction brought about by the polarization effect of conduction electrons, which may lead to magnetic phase at sufficiently high impurity concentrations.¹²⁻¹⁵ The complete theory should discuss the dynamics of the impurity spins coupled with the conduction electrons. Moreover, at higher concentrations the proper averaging procedure over the different impurity configurations taken into account by the partial square vertex part Φ should lead to significant corrections. All that one may expect is that the qualitative fea-

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tures of the presented theoretical calculation should agree reasonably well with the experimental results at low concentrations of paramagnetic impurities.

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