The influence of the energy gap of a superconductor on its inelastic neutron scattering parameters: nonequilibrium effects

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In this paper we determine the differential cross-section with respect to energy and angle for neutron scattering by rare-earth ions (REI) in superconductors. We show that a nonequilibrium distribution of electrons induced by an external AC electromagnetic field leads to resonant growth of the neutron scattering cross-section and to increased transition line widths for energies lying within the region $2\Delta(T) \leqslant \delta_{ij} \leqslant 2\Delta(T) + \omega_0$ (where $\delta_{ij} = E_i - E_j$, E_i, E_j are the energy levels of the REI in the crystal field, $2\Delta(T)$ is the energy gap of the superconductor at a temperature T, and ω_0 is the frequency of the electromagnetic field). The estimates that we have obtained for the high-temperature superconductor $Tm_{0.1}Y_{0.9}Ba_2Cu_3O_{6.9}$ show that this resonant growth can increase the integrated scattering intensities by as much as 20% and the line widths by as much as 12.8%.

1. INTRODUCTION

Experimental investigations of the properties of compounds such as $La_{1-x}Tb_xAl_2$ (Ref. 1), $Tm_{0.1}Y_{0.9}Ba_2Cu_3O_{6.9}$ (Ref. 2), $ErBa_2Cu_3O_7$ (Ref. 3) and others based on inelastic neutron scattering have shown that the line widths of transitions between crystal-field levels of the rare-earth ions change considerably when these compounds enter the superconducting state. The theoretical calculations of Refs. 4 and 5, which include both ordinary relaxation processes and recombination effects associated with the breaking of Cooper pairs for $\delta_{ij} \gtrsim 2\Delta(T)$, give a satisfactory description of the experimental results. However, it is quite difficult to determine values of the energy gap by using data from measurements of the temperature dependence of the transition line widths. The primary reason for this is the fact that, although there may be a considerable variation in the width of a given transition line with temperature starting near $T = T_c$ (T_c is the superconducting transition temperature), in most cases a plot of the linewidth versus T shows at most a change in slope for $2\Delta(T) = \delta_{ij}$ (Refs. 1, 2). Nevertheless, theoretical considerations suggest that when the edges of the energy gap are sufficiently sharp, the probability for crossed quantum transitions between the conduction-electron and rare-earth-ion (REI) systems should be resonantly enhanced when $2\Delta(T) = \delta_{ij}$. Such a change of the transition probability ought to influence the occupation of the REI levels and their lifetimes, and consequently the parameters for inelastic neutron scattering; these effects can be resonant when, for example, the system of conduction electrons is transferred to a nonequilibrium state.

From an experimental point of view, the most desirable situation of this kind is one in which a resonant variation of some measurable physical quantity occurs at those temperatures T for which the equation $2\Delta(T) = \delta_{ij}$ is satisfied. Note that the problem of determining the parameters of neutron scattering by electrons in the magnetic shell of the ions is directly related to the more general problem of studying the kinetics of the occupation of the REI levels in nonequilibrium superconducting compounds. More specifically, the dynamic response function that determines the neutron scattering cross-section of a system under the influence of an external nonequilibrium source will be expressed in terms of the nonequilibrium distribution function of the electrons and REI. However, in order to find the distribution function it is necessary to determine kinetic equations for the electron and ion subsystems; in the general case this also requires that we determine the coupled kinetics of the occupation of the REI levels and the conduction electrons.

It is certainly true that the solution of coupled kinetic equations for ions and electrons constitutes a complicated problem. In this paper we will simplify the problem by first imposing the limitation of small REI concentrations; this allows us to neglect the inverse effect of the ionic subsystem on those quasiparticle excitations that are "heated" by the external field. At this time, the problem of the kinetics of the electron subsystem can be regarded as rather well understood.[6,7] One of the most powerful methods used to study electron kinetics is the method of Green's functions integrated with respect to energy,[8,9] which is based on the diagram technique for nonequilibrium systems.[10] In this paper we also assume that in constructing the kinetic equations for the ion system we can describe the electronic component entering into the latter by using Green-Gor'kov-Eilenberger-Eliashberg functions.

Based on the above assumptions, in this paper we have made an attempt to study the kinetics of the occupation of the REI levels in nonequilibrium superconductors. The quantity we have chosen to study is the cross section for inelastic neutron scattering by the rare-earth elements. The system is driven out of equilibrium by a variable electromagnetic field which acts continuously on the superconductor; the frequency of this field satisfies $\omega_0 < 2\Delta(T)$, i.e., it cannot give rise to the creation of quasiparticles above the energy gap.

2. STATEMENT OF THE PROBLEM

The neutron scattering cross section reflects the dynamic properties of the target at various frequencies and wavelengths. In connection with this, based on the fluctuation-dissipation theorem, the response of a system to a weak perturbation that depends on frequency and wave vector is usually described by using the dynamic susceptibility.[11,12]

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Thus, for the differential scattering cross section of unpolarized neutrons with respect to energy and angle we have[12]

$$\frac{\partial^2\sigma}{\partial\Omega\partial E'}=\frac{k'}{4k}\frac{\gamma e^2}{m_0c^2}\left\{\frac{1}{2}gF(Q)\exp[W(Q)]\right\}^2$$
$$\times\frac{1}{2\pi}\int_{-\infty}^{+\infty}dt\exp(-i\omega t)$$
$$\times\sum_{\mathbf{l},\mathbf{l}'}\exp[i\mathbf{Q}(\mathbf{l}-\mathbf{l}')]\{\langle\hat{S}_l^-(0)\hat{S}_{l'}^+(t)\rangle+\langle\hat{S}_l^+(0)\hat{S}_{l'}^-(t)\rangle\},\tag{1}$$

where

$$\mathbf{Q}=\mathbf{k}-\mathbf{k}';\quad \hbar\omega=\frac{1}{2m_0}(k^2-k'^2)=E-E';$$

$$\frac{\gamma e^2}{m_0c^2}=-0{,}54\cdot10^{-12}\text{ cm},$$

$\mathbf{k}$, $\mathbf{k}'$ are the wave vectors of the neutron in its initial and final states, respectively, $W(Q)$ is the Debye-Waller factor, and $F(Q)$ is a form factor. The correlation functions of the localized spin operators $\hat{S}_l^-$ and $\hat{S}_l^+$ in Eq. (1) are expressed in terms of the dynamic susceptibility as follows:

$$S^{1(2)}(\omega)=\frac{1}{2\pi}\int_{-\infty}^{+\infty}dt\exp(-i\omega t)\langle\hat{S}^{-(+)}(0)\hat{S}^{+(-)}(t)\rangle,$$
$$S^i(\omega)=-\frac{1}{\pi}\frac{1}{1-\exp(-\beta\omega)}\operatorname{Im}\chi_i(i\omega),\quad i=1,2.\tag{2}$$

In order to calculate the susceptibility in Eq. (2), we introduce the Hamiltonian of the problem:

$$\hat{\mathcal{H}}=\hat{\mathcal{H}}_0+\hat{\mathcal{H}}_{int},$$
$$\hat{\mathcal{H}}_0=\sum_\sigma\int dr\,\psi_\sigma^+(\mathbf{r})\left[-\frac{1}{2m}\left(\nabla-i\frac{e}{c}\mathbf{A}\right)^2\right]\psi_\sigma(\mathbf{r})$$
$$-|V|\int dr\,\psi_+^+(r)\psi_-^+(r)\psi_-(r)\psi_+(r)+\sum_{l,i}(\lambda+\delta_i)a_i^+(l)a_i(l),$$
$$\hat{\mathcal{H}}_{int}=\sum_{lij\sigma\sigma'}M_{\sigma\sigma'}^{ij}a_i^+(l)a_j(l)\psi_\sigma^+(\mathbf{r}_l)\psi_{\sigma'}(\mathbf{r}_l)\tag{3}$$

in which $\hat{\mathcal{H}}_0$ describes the free motion of conduction electrons and REI in the crystal field of the superconductor while $\hat{\mathcal{H}}_{int}$ is the operator which describes the interaction of the conduction electrons with the REI. Here $\psi_\sigma^+(r)$, $\psi_\sigma(r)$ are field operators for electrons, $\mathbf{A}$ is the vector potential of the electromagnetic field, $M_{\sigma\sigma'}^{ij}$ is the matrix element for a transition between states i and j with energies δ_i and δ_j of the REI, and $a_i^+(l)$, $a_i(l)$ are spin fermion operators, which are connected with the usual spin operators $\hat{\mathbf{S}}_l$ by the relation[13]

$$\hat{\mathbf{S}}_l=\sum_{ij}a_i^+(l)\hat{\mathbf{S}}_{ij}a_j(l).\tag{4}$$

λ plays the role of a chemical potential, which "freezes out" the unphysical states in the limit[14]

$$\prod_l\lim_{\lambda(l)\to\infty}\left\{\frac{\exp[\beta\lambda(l)]}{\sum_i\exp(-\beta\delta_i)}[\ldots]\right\}.\tag{5}$$

Thus, our task is to determine the dynamic response function $\chi(i\omega)=\chi_1(i\omega)+\chi_2(i\omega)$, taking into account the definitions (4) and (5), where

$$\chi_1(i\omega)=\lim_{\lambda\to\infty}\frac{ce^{\beta\lambda}}{Z}\chi_{ij}(\omega)$$
$$=\lim_{\lambda\to\infty}\frac{ce^{\beta\lambda}}{Z}\langle a_i^+(t)a_j(t)a_j^+(t')a_i(t')\rangle_\omega,$$
$$\chi_2(i\omega)=\lim_{\lambda\to\infty}\frac{ce^{\beta\lambda}}{Z}\chi_{ji}(\omega)$$
$$=\lim_{\lambda\to\infty}\frac{ce^{\beta\lambda}}{Z}\langle a_j^+(t)a_i(t)a_i^+(t')a_j(t')\rangle_\omega,$$
$$Z=\exp(-\beta\delta_i)+\exp(-\beta\delta_j);\tag{6}$$

here Z is the partition function[1)] and c is the REI concentration in the sample. Because we have assumed that $c\ll1$ and are neglecting ion–ion interactions, in carrying out the calculation we will assume that the change in the density of electron levels caused by scattering of the conduction electrons by REI is small and therefore does not lead to any appreciable change in the superconducting order parameter.

3. KINETIC EQUATION

The most general method used in microscopic investigations of nonequilibrium systems is the method of Keldysh.[10] In this method, the equation for the Green's function of the REI takes the form

$$\left\{i\hat{\sigma}_z\frac{\partial}{\partial t}-\lambda-\delta_i-\hat{\Sigma}_i\right\}\hat{D}_i=\hat{1}\delta(t-t')\delta_{ll'},$$
$$\hat{D}_i=\hat{D}_i(l,t;l',t')=\begin{pmatrix}D_i^R & D_i\\ 0 & D_i^A\end{pmatrix};\tag{7}$$
$$\hat{\Sigma}_i=\begin{pmatrix}\Sigma_i^R & \Sigma_i\\ 0 & \Sigma_i^A\end{pmatrix};\quad \hat{\sigma}_z=\begin{pmatrix}1 & 0\\ 0 & -1\end{pmatrix},$$

where D_i^R, D_i^A are the advanced and retarded Green's functions, which are expressed in terms of the Green-Keldysh function in the usual way:

$$D_i^R=D_i^{--}-D_i^{-+};\quad D_i^A=D_i^{--}-D_i^{+-};\quad D_i=D_i^{--}+D_i^{++},$$
$$iD_i^{--}(l,t;l't')=\begin{cases}\langle a_i(lt)a_i^+(l't')\rangle, & t>t',\\ -\langle a_i^+(l't')a_i(lt)\rangle, & t<t',\end{cases}$$
$$iD^{-+}(lt,l't')=-\langle a_i^+(l't')a_i(lt)\rangle,\tag{8}$$
$$iD^{+-}(lt,l't')=\langle a_i(lt)a_i^+(l't')\rangle.$$

The following identities are obtained for the self-energy parts:[15]

$$\Sigma_i^R=\Sigma_i^{--}+\Sigma_i^{-+}=-\Sigma_i^{++}-\Sigma_i^{+-},\quad \Sigma_i=\Sigma_i^{--}+\Sigma_i^{++},$$
$$\Sigma_i^A=\Sigma_i^{--}+\Sigma_i^{+-}=-\Sigma_i^{++}-\Sigma_i^{-+}.\tag{9}$$

Using Eq. (7) we can obtain quasiclassical equations for the Green's function[9]

$$\hat{\sigma}_z\frac{\partial}{\partial t}\hat{D}_{itt'}+\frac{\partial}{\partial t'}\hat{D}_{itt'}\hat{\sigma}_z+i\int_{-\infty}^{+\infty}dt_1\{\hat{\Sigma}_{itt_1}\hat{D}_{it_1t'}-\hat{D}_{itt_1}\hat{\Sigma}_{it_1t'}\}=0,\tag{10}$$
$$\hat{D}_{itt'}=\hat{D}_i(l,t,t')\delta_{ll'},\quad \hat{\Sigma}_{itt_1}=\Sigma_i(l,t,t_1)\delta_{ll'}.$$

Assuming from here on that the functions entering into Eq. (10) vary rapidly with $\tau = t - t_1$, we Fourier transform Eq. (10) with respect to this difference variable. Eventually we obtain for the $(-+)$ component

$$\frac{\partial}{\partial t} n_i(\lambda, t) = i n_i(\lambda, t) \Sigma_i^{+-}(\lambda+\delta_i, t) - i[n_i(\lambda, t)-1]\Sigma_i^{-+}(\lambda+\delta_i, t). \quad (11)$$

In obtaining Eq. (11) we have used the functions D_i^{-+}, D_i^{+-} in the following form:

$$D_i^{-+}(\omega, t) = 2\pi i n_i(\lambda, t)\delta(\omega-\lambda-\delta_i),$$
$$D_i^{+-}(\omega, t) = 2\pi i[n_i(\lambda, t)-1]\delta(\omega-\lambda-\delta_i), \quad (12)$$

where in equilibrium $n_i(\lambda,t)$ reduces to the function $n_i^0(\lambda)$:

$$n_i^0(\lambda) = [e^{\beta(\lambda+\delta_i)}+1]^{-1}. \quad (13)$$

Relations (12) are obtained by standard methods (see Ref. 15) based on the Hamiltonian (3) and the definition of the functions D_i^{-+}, D_i^{+-} in Eq. (8).

Equation (11) is our basis for obtaining kinetic equations for the occupation of the REI levels. To do this it is first necessary to find explicit forms for the components of the self-energy parts $\Sigma_i^{-+}(\lambda+\delta_i)$ and $\Sigma_i^{+-}(\lambda+\delta_i)$ and, secondly, to annihilate the unphysical states in Eq. (11) by taking the limit with respect to the parameter λ given in Eq. (5).

Let us introduce the electron Green-Keldysh functions in the usual way:

$$(G)_\beta^{\alpha,mn}(1,1') = -i\langle T\psi_\beta(1m)\psi^\alpha(1'n)\rangle,$$

where $(1) = (r_1,t_1)$; the indices m and n indicate which of the two branches of the Keldysh contour the time coordinate will follow.[10] Once we have introduced the single-particle functions for conduction electrons, we can write down expressions for the self-energy parts $\hat{\Sigma}_i$ in explicit form. Thus, for the components Σ_i^{-+} and Σ_i^{+-}, we obtain to second order in $\hat{\mathscr{H}}_{int}$ the following:

$$\Sigma_i^{+-,(-+)}(lt_1, l't_2) = -\frac{1}{2}\sum_{j\sigma\sigma'}[M_{\sigma,\sigma'}^{ij} M_{\sigma',\sigma}^{ji} G_{\sigma',\sigma'}^{+-(-+)}(lt_1, l't_2) G_{\sigma,\sigma}^{-+(+-)}(l't_2, lt_1)$$
$$- M_{\sigma,\sigma'}^{ij} M_{-\sigma,-\sigma'}^{ji} F_{\sigma,-\sigma}^{+-(-+)+}(lt_1, l't_2) F_{-\sigma',\sigma'}^{-+(+-)}$$
$$\times (l't_2, lt_1)] D_j^{+-(-+)}(lt_1, l't_2), \quad (14)$$

where G and F are, respectively, the normal and anomalous Green's functions of the superconductor.

After Fourier-transforming Eq. (14) with respect to the time difference and passing from the total Green's function to the Green-Gor'kov-Eilenberger-Eliashberg function integrated over energy,[8,9] we obtain for Σ_i^{-+} and Σ_i^{+-}

$$\Sigma_i^{-+}(\lambda+\delta_i, t) = -\pi i n_j(\lambda, t)\rho^2 \sum_{\sigma\sigma'}\Big\{\int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty} L_{ij\sigma\sigma'}^{-}(\varepsilon_1, \varepsilon_2) f(\varepsilon_1)[1-f(\varepsilon_2)]$$
$$\times \delta(\varepsilon_1-\varepsilon_2-\delta_{ij})\, d\varepsilon_1\, d\varepsilon_2 + \int_0^{\infty}\int_0^{\infty} L_{ij\sigma\sigma'}^{+}(\varepsilon_1, \varepsilon_2) f(\varepsilon_1) f(\varepsilon_2)\delta(\varepsilon_1+\varepsilon_2-\delta_{ij})\, d\varepsilon_1\, d\varepsilon_2\Big\}, \quad (15)$$

$$\Sigma_i^{+-}(\lambda+\delta_i, t) = \pi i[1-n_j(\lambda, t)]\rho^2 \sum_{\sigma\sigma'}\Big\{\int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty} L_{ij\sigma\sigma'}^{-}(\varepsilon_1, \varepsilon_2)[1-f(\varepsilon_1)] f(\varepsilon_2)$$
$$\times \delta(\varepsilon_1-\varepsilon_2-\delta_{ij})\, d\varepsilon_1\, d\varepsilon_2 + \int_0^{\infty}\int_0^{\infty} L_{ij\sigma\sigma'}^{+}(\varepsilon_1, \varepsilon_2)[1-f(\varepsilon_1)][1-f(\varepsilon_2)]$$
$$\times \delta(\varepsilon_1+\varepsilon_2-\delta_{ij})\, d\varepsilon_1\, d\varepsilon_2\Big\},$$

where

$$L_{ij\sigma\sigma'}^{\pm}(\varepsilon_1, \varepsilon_2) = \frac{M_{\sigma,\sigma'}^{ij} M_{\sigma',\sigma}^{ji}\varepsilon_1\varepsilon_2 \pm M_{\sigma,\sigma'}^{ij} M_{-\sigma,-\sigma'}^{ji}\Delta^2}{(\varepsilon_1^2-\Delta^2)^{1/2}(\varepsilon_2^2-\Delta^2)^{1/2}}$$
$$\times \theta(\varepsilon_1^2-\Delta^2)\theta(\varepsilon_2^2-\Delta^2);$$

here ρ is the density of states of conduction electrons at the Fermi surface in the normal state, and $f(\varepsilon)$ is the nonequilibrium quasielectron distribution function. We assume that the usual symmetry of the electron and hole branches of the distribution function for the excitations obtains, and that this symmetry also holds in the nonequilibrium case, i.e., under excitation by the high-frequency electromagnetic field.[16] In what follows we do not take into account the dependence of the matrix elements $M_{\sigma\sigma'}^{ij}$ on the components σ, σ' of the electron spin, and therefore we assume that

$$M_{ij} = M_{\sigma,\sigma'}^{ij} = (M_{\sigma',\sigma}^{ji})^* = (M_{-\sigma,-\sigma'}^{ji})^*. \quad (16)$$

Nevertheless, we note that the validity of the last equality in Eq. (16) is a question that requires a special investigation. It is possible that in certain compounds the question of the spin dependence of $M_{\sigma\sigma'}^{ij}$ can turn out to be important, because the symmetry properties of these matrix elements, as is clear from Eq. (15), are directly related to the sign of the coherence factor.

Thus, by using Eqs. (11), (15), and (5) it is easy to obtain the kinetic equations for the occupation of the REI levels:

$$\frac{d}{dt} n_i(t) = 2\pi |M_{ij}|^2\rho^2 \Big\{\int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty} \frac{(\varepsilon_1\varepsilon_2-\Delta^2)\theta(\varepsilon_1^2-\Delta^2)\theta(\varepsilon_2^2-\Delta^2)}{(\varepsilon_1^2-\Delta^2)^{1/2}(\varepsilon_2^2-\Delta^2)^{1/2}}$$
$$\times [n_j(t) f(\varepsilon_1)[1-f(\varepsilon_2)] - n_i(t) f(\varepsilon_2)[1-f(\varepsilon_1)]]$$
$$\times \delta(\varepsilon_1-\varepsilon_2-\delta_{ij})\, d\varepsilon_1\, d\varepsilon_2$$
$$+ \int_0^{\infty}\int_0^{\infty} \frac{(\varepsilon_1\varepsilon_2+\Delta^2)\theta(\varepsilon_1^2-\Delta^2)\theta(\varepsilon_2^2-\Delta^2)}{(\varepsilon_1^2-\Delta^2)^{1/2}(\varepsilon_2^2-\Delta^2)^{1/2}}[n_j(t) f(\varepsilon_1) f(\varepsilon_2)$$
$$- n_i(t)[1-f(\varepsilon_1)][1-f(\varepsilon_2)]]\delta(\varepsilon_1+\varepsilon_2-\delta_{ij})\, d\varepsilon_1\, d\varepsilon_2\Big\}, \quad (17)$$

where $n_i(t)$, $n_j(t)$ are nonequilibrium occupation numbers for the ith and jth ionic levels, respectively. If we pass to equilibrium (or turn off the source of nonequilibrium excitation) we find

$$n_i(t)\to n_i^0=Z^{-1}e^{-\beta\delta_i}. \qquad (18)$$

As we should expect, the right-hand side of the kinetic equation (17), which is the collision integral, reduces to zero when we replace the distribution functions n and f by their equilibrium values.

The problem of finding the electronic distribution function $f(\varepsilon)$ of a superconductor that is subjected to electromagnetic radiation has been discussed in detail in Refs. 6 and 17. Considerable progress has been made in investigating the nonlinear electrodynamics of superconductors based on the model proposed by Eliashberg with a phonon heat bath.[6,7] According to this model, the coupled kinetic equations for electrons and phonons decouple for the case of sufficiently thin films, in which the nonequilibrium phonons that are generated by the field succeed in leaving the film without giving rise to any change in the stationary distribution of conduction electrons. Moreover, it is possible to represent electron kinetic equations that are initially given in terms of Green-Gor'kov-Eilenberger-Eliashberg functions integrated over energy in the form of Boltzmann equations. This situation is realized for the case of a system with a sufficient number of nonmagnetic impurities. By taking into account the limitations mentioned above, and assuming that the external electromagnetic field with frequency ω_0 is incident perpendicular to a sample film, so that the vector potential $\mathbf{A}$ lies in the plane of the film, and that the film thickness is smaller than the penetration depth of the field, the authors of Refs. 6 and 18 obtained the following expression for $f(\varepsilon)$:

$$f(\varepsilon)-f_0(\varepsilon)=f_1(\varepsilon)$$

$$=\alpha\tau_{en}\frac{\omega_0}{2T}\,\mathrm{ch}^{-2}\frac{\varepsilon}{2T}\,[U_{\varepsilon,\varepsilon-\omega_0}-U_{\varepsilon+\omega_0,\varepsilon}]\frac{(\varepsilon^2-\Delta^2)^{1/2}}{\varepsilon},\quad \varepsilon>\Delta, \qquad (19)$$

where

$$U_{\varepsilon,\varepsilon-\omega_0}=\frac{\varepsilon(\varepsilon-\omega_0)+\Delta^2}{(\varepsilon^2-\Delta^2)^{1/2}((\varepsilon-\omega_0)^2-\Delta^2)^{1/2}}\theta(\varepsilon-\omega_0-\Delta),$$

$$\alpha=D\left(\frac{l}{c}\right)^2\mathbf{A}_{\omega_0}\mathbf{A}_{-\omega_0}, \qquad (20)$$

$D=\frac{1}{3}v_F^2\tau$ is the diffusion coefficient and $\mathbf{A}_{\omega_0}$ is the Fourier component of the vector potential at frequency ω_0.

Substituting Eqs. (19) and (20) into the right side of the kinetic equation (17) and setting it equal to zero, we obtain for the population of the REI levels

$$n_j-n_i=n_j^0-n_i^0-2n, \qquad (21)$$

where

$$n=\frac{n_j^0\beta_1-n_i^0\alpha_1}{\alpha_0(1+e^{-\beta\delta_{ij}})}; \qquad (22)$$

n_i^0, n_j^0 are the equilibrium populations determined by Eq. (18). The quantities α_0, α_1, and β_1 are given in the Appendix.

4. DYNAMIC RESPONSE

Using the Keldysh method[10] the equations for the nonequilibrium susceptibility (6) can be written in the form

$$\left(i\frac{\partial}{\partial t}-\delta_{ij}\right)\hat{\chi}_{ji}(t,t',t)=\delta(t-t')[\hat{D}_j(t',t)-\hat{D}_i(t,t')]$$

$$-\int dt_1[\hat{\Sigma}_j(t,t_1)\hat{\chi}_{ji}(t_1,t',t)-\hat{\chi}_{ji}(t,t',t_1)\hat{\Sigma}_i(t_1,t)], \qquad (23)$$

$$\hat{\chi}_{ji}=\begin{pmatrix}\chi_{ji}^R & \chi_{ji}\\ 0 & \chi_{ji}^A\end{pmatrix}. \qquad (24)$$

The equations for $\hat{\chi}_{ij}(t,t',t)$ are obtained from Eq. (23) by exchanging the positions of the subscripts i and j. The matrices $\hat{D}_i$, $\hat{\Sigma}_i$ are determined in Eq. (7). The following relations can be derived for the components $\hat{\chi}_{ij}$ from Eq. (24); note that these hold for the single-particle Green-Keldysh function as well:

$$\chi_{ji}^R=\chi_{ji}^{--}-\chi_{ji}^{-+},\quad \chi_{ji}^A=\chi_{ji}^{--}-\chi_{ji}^{+-},\qquad (25)$$
$$\chi_{ji}=\chi_{ji}^{--}+\chi_{ji}^{++}.$$

After passing to a mixed representation in Eq. (23) and taking the Fourier transform with respect to the time difference, we obtain for the components of Eq. (12)

$$(\omega-\delta_{ij})\chi_{ji}(t,\omega)$$

$$=\int\frac{d\omega_1}{2\pi}\{D_j(t,\omega_1)-D_i(t,\omega_1+\omega)-[\Sigma_j^R(t,\omega_1+\omega)$$

$$-\Sigma_i^A(t,\omega_1)]\chi_{ji}(t,\omega_1,\omega_1+\omega)+\chi_{ji}^R(t,\omega_1,\omega_1+\omega)\Sigma_i(t,\omega_1)$$

$$-\Sigma_j(t,\omega_1+\omega)\chi_{ji}^A(t,\omega_1,\omega_1+\omega)\}. \qquad (26)$$

By using Eqs. (6), (9), (15), and the condition of thermodynamic equilibrium of the system consisting of the superconductor plus external electromagnetic field, after some uncomplicated mathematical transformations we find from Eq. (26)

$$\chi_1(i\omega)=\frac{2c(n_i-n_j)}{\omega+\delta_{ij}-\Sigma_j^{+-}(\omega=\lambda+\delta_j)|_{\lambda\to+\infty}}, \qquad (27)$$

$$\chi_2(i\omega)=\frac{2c(n_j-n_i)}{\omega-\delta_{ij}-\Sigma_i^{+-}(\omega=\lambda+\delta_i)|_{\lambda\to+\infty}}, \qquad (28)$$

where

$$\Sigma_i^{+-}(\omega=\lambda+\delta_i)|_{\lambda\to+\infty}=2\pi i|M_{ij}|^2\rho^2(\alpha_0+\alpha_1),$$
$$\Sigma_j^{+-}(\omega=\lambda+\delta_j)|_{\lambda\to+\infty}=2\pi i|M_{ij}|^2\rho^2(\alpha_0e^{-\beta\delta_{ij}}+\beta_1);$$

n_j-n_i is determined in Eq. (21). The quantities α_0, α_1, and β_1 are given by Eqs. (A1)–(A3) of the Appendix.

Finally, taking into account Eqs. (1), (2), (27), and (28), the equation for the differential cross section with respect to energy and angle for scattering of neutrons by REI takes the form

$$\frac{\partial^2\sigma}{\partial\Omega\,\partial E'}=\frac{k}{4k'}\frac{\gamma e^2}{m_0c^2}\left\{\frac{1}{2}gF(Q)\exp[W(Q)]\right\}^2$$

$$\times\frac{2c}{\pi}\frac{n_j-n_i}{1-\exp(-\beta\omega)}\frac{2\delta_{ij}\omega(\Gamma+K)+(\omega^2+\delta_{ij}^2)(\Gamma-K)}{(\omega^2-\delta_{ij}^2)^2+[\omega(\Gamma+K)+\delta_{ij}(\Gamma-K)]^2}$$

where

$$\Gamma = 2\pi |M_{ij}|^2 \rho^2 (\alpha_0 + \alpha_1),$$
$$K = 2\pi |M_{ij}|^2 \rho^2 (\alpha_0 e^{-\beta \delta_{ij}} + \beta_1). \tag{29}$$

5. DISCUSSION OF RESULTS

As is clear from Eqs. (A1)–(A3), the quantities α_1 and β_1 vary significantly in the region of energies determined by the condition

$$2\Delta(T) \leqslant \delta_{ij} \leqslant 2\Delta(T) + \omega_0. \tag{30}$$

Note that when condition (30) is fulfilled the quantity n given by Eqs. (21) and (22) becomes negative. At first glance this leads to a paradoxical result: when the superconductor is "heated" by an external electromagnetic field a cooling of the REI system occurs, i.e., the occupation of the ground state level of an ion in the nonequilibrium superconductor becomes larger than its value in the absence of the field. However, this effect has a simple explanation: in the absence of electromagnetic pumping, for $\delta_{ij} \sim 2\Delta(T)$ "crossed" quantum transitions occur between crystal-field levels of the REI system and across the energy gap in the system of conduction electrons. The numbers of self-induced transitions "upward" and "downward" with respect to the energy are the same under equilibrium conditions. When the electromagnetic field with frequency $\omega_0 < 2\Delta(T)$ is switched on, a redistribution occurs of the quasiparticles that are located above the energy gap into regions of higher energies; because energy states directly above the energy gap are thereby exposed, this leads to an enhancement of the upward transitions across to the energy gap in the system of conduction electrons, and correspondingly to an enhancement of the downward transitions to the ground state level of the REI system. The new thermodynamic equilibrium of the system of superconductor plus AC electromagnetic field will be characterized by an increase in the population differences of the ground and excited levels of the REI $n_j - n_i$ above their values $n_j^0 - n_i^0$ which follow from the Boltzmann distribution.

The situation described above is in a certain sense similar to the "phonon deficit" effect investigated by Gulyan and Zharkov:[18] according to these authors, a superconducting thin film that is placed in a heat bath and subject to an external electromagnetic field with frequency $\omega_0 < 2\Delta$ can effectively absorb phonons within a certain spectral interval from the heat bath.

According to Eq. (29), the increase in $n_j - n_i$ should change the scattering cross-section; in addition, by virtue of the coupling of the parameters Γ and K to α_1 and β_1 as given by Eq. (29), it should also change the width of the transition lines. In order to estimate the magnitude of this effect, we use experimental data from neutron-scattering studies of the high-temperature superconductor $Tm_{0.1}Y_{0.9}Ba_2Cu_3O_{6.9}$ (see Ref. 2). In this compound, for which $T_c = 92 \pm 2$ K, the ground-state $^3H_6$ multiplet of the Tm^{3+} ion, which is split by an electronic crystal field of orthorhombic symmetry, is characterized by the following low-lying levels: the ground state level Γ_3 and the two lowest excited states Γ_4 and Γ_2 with energies 11.8 and 14.2 meV, respectively, and by dipole-allowed transitions from the ground state level. With regard to the case of a two-level system which we are discussing here, we can assume a value $\delta_{ij} = 11.8$ meV for the transition Γ_3–Γ_4. This quantity yields an estimate of $2\Delta(T) = \delta_{ij} = 136.84$ K, since according to Ref. 2 we have $2\Delta(0) > 14.8$ eV. Assuming values for the other parameters that agree with analogous estimates that can be found in the literature [$\omega_0 = 1\ \text{K} = 2\pi \times 0.21 \times 10^{10}\ \text{sec}^{-1}$ (see Ref. 19), $B_{\omega_0} = 2 \times 10^{-4}$ Oe for the magnetic induction of the electromagnetic field,[20] $\tau_{en} = 10^{-9}$ sec (see Ref. 21), $v_F = 2 \times 10^6$ m/sec, $\tau = 2 \times 10^{-13}$ sec], let us plot the dependence of $n_j - n_i$ on $2\Delta(T)$ in the energy region determined by condition (30) (see the figure).

Introducing the notation

$$s = \frac{\partial^2 \sigma}{\partial \Omega\, \partial E'}, \quad 2\Delta(T) \leqslant \delta_{ij} \leqslant 2\Delta(T) + \omega_0, \tag{31a}$$

$$s_0 = \frac{\partial^2 \sigma}{\partial \Omega\, \partial E'}, \quad \delta_{ij} < 2\Delta(T), \tag{31b}$$

we estimate the ratio s/s_0 for $\omega = \delta_{ij}$, i.e., at the center of the scattering line:

$$s/s_0 \sim \frac{n_j - n_i}{n_j^0 - n_i^0} \frac{\Gamma_0}{\Gamma}, \quad \Gamma_0 = 2\pi |M_{ij}|^2 \rho^2 \alpha_0, \tag{32}$$

where $\Gamma - \alpha_0 + \alpha_1$ is the line width for cases (31a) and (31b), respectively. Estimates based on Eqs. (A1)–(A3) and Eq. (32), using the parameters presented above, give

$$\Gamma/\Gamma_0 = \frac{\alpha_0 + \alpha_1}{\alpha_0} \sim 1{,}128, \qquad \frac{n_j - n_i}{n_j^0 - n_i^0} \sim 1{,}2, \qquad s/s_0 \sim 1{,}064. \tag{33}$$

It follows from Eq. (33) that when condition (30) is fulfilled the following quantities increase in a resonant fashion: the line width (Γ/Γ_0) increases by 12.8%, the integrated scattering intensity ($\sim n_j - n_i/n_j^0 - n_i^0$) increases by 20%, and the maximum of the scattering cross section (s/s_0) increases by 6.4%. Of course, all of these estimates are conditional, since as the intensity of the source that drives the system out of equilibrium varies the ratios in Eq. (33) can increase significantly until inverse saturation occurs, i.e., at $n_j - n_i = 1$. The study of nonequilibrium kinetics under conditions of supercooling of the ion system, which certainly is an interesting problem, lies outside the framework of this paper since it can be addressed only by taking into account such distinctive features as the change in the density of electronic levels and the superconducting order parameter brought about an intense electromagnetic field, the appearance of

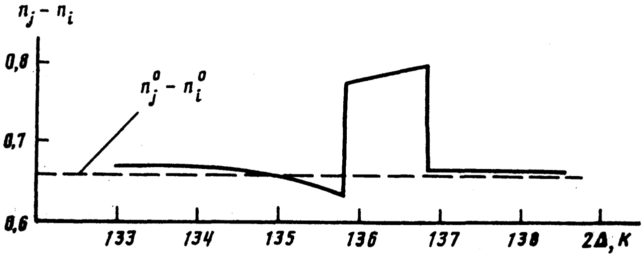

FIG. 1. Dependence of the population differences of REI levels on the magnitude of the energy gap obtained according to Eq. (21). The values of the parameters are presented in the text. The dashed line denotes the equilibrium population difference calculated for $T = 86$ K.

shock ionization effects,[22] etc.

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APPENDIX

Let us write the following obvious relations for the occupation of a two-level REI system:

$$n_i=n_i^0+n, \quad n_j=n_j^0-n,$$

from which, in particular, there follows:

$$n_i+n_j=n_i^0+n_j^0, \quad n_j-n_i=n_j^0-n_i^0-2n.$$

After linearizing the kinetic Eq. (17) with respect to the amplitude of the external electromagnetic field, we obtain for n

$$n=\frac{n_j^0\beta_1-n_i^0\alpha_1}{\alpha_0(1+e^{-\beta\delta_{ij}})},$$

where

$$\alpha_0=\int_\Delta^\infty\int_\Delta^\infty L^-(\varepsilon_1,\varepsilon_2)f_0(\varepsilon_2)[1-f_0(\varepsilon_1)]\delta(\varepsilon_1-\varepsilon_2-\delta_{ij})\,d\varepsilon_1\,d\varepsilon_2$$
$$+\int_\Delta^\infty\int_\Delta^\infty L^+(\varepsilon_1,\varepsilon_2)[1-f_0(\varepsilon_1)][1-f_0(\varepsilon_2)]\delta(\varepsilon_1+\varepsilon_2-\delta_{ij})\,d\varepsilon_1\,d\varepsilon_2, \qquad \text{(A1)}$$

$$\alpha_1=\int_\Delta^\infty\int_\Delta^\infty L^-(\varepsilon_1,\varepsilon_2)f_1(\varepsilon_2)\delta(\varepsilon_1-\varepsilon_2-\delta_{ij})\,d\varepsilon_1\,d\varepsilon_2$$
$$-\int_\Delta^\infty\int_\Delta^\infty L^+(\varepsilon_1,\varepsilon_2)\{[1-f_0(\varepsilon_1)]f_1(\varepsilon_2)+f_1(\varepsilon_1)[1-f_0(\varepsilon_2)]\}$$
$$\times\delta(\varepsilon_1+\varepsilon_2-\delta_{ij})\,d\varepsilon_1\,d\varepsilon_2, \qquad \text{(A2)}$$

$$\beta_1=\int_\Delta^\infty\int_\Delta^\infty L^-(\varepsilon_1,\varepsilon_2)f_1(\varepsilon_1)\delta(\varepsilon_1-\varepsilon_2-\delta_{ij})\,d\varepsilon_1\,d\varepsilon_2$$
$$+\int_\Delta^\infty\int_\Delta^\infty L^+(\varepsilon_1,\varepsilon_2)[f_0(\varepsilon_1)f_1(\varepsilon_2)+f_1(\varepsilon_1)f_0(\varepsilon_2)]$$
$$\times\delta(\varepsilon_1+\varepsilon_2-\delta_{ij})\,d\varepsilon_1\,d\varepsilon_2, \qquad \text{(A3)}$$

$$L^\pm(\varepsilon_1,\varepsilon_2)=\frac{\varepsilon_1\varepsilon_2\pm\Delta^2}{(\varepsilon_1^2-\Delta^2)^{1/2}(\varepsilon_2^2-\Delta^2)^{1/2}}\theta(\varepsilon_1^2-\Delta^2)\theta(\varepsilon_2^2-\Delta^2).$$

The integrals (A1)–(A3) can be calculated by using the mean-value theorem Eq. (23), which separates out the slowly-varying part of the function under the integral sign. Substituting the explicit form of the correction to the electron distribution function $f_1(\varepsilon)$ with respect to the field into Eqs. (A1)–(A3), to the accuracy of the mean-value theorem we obtain

$$\alpha_0=\left\{\frac{e^{\beta\delta_{ij}}}{e^{\beta\delta_{ij}}-1}\left[\left(\delta_{ij}-2\Delta+2T\ln\frac{e^{\beta\delta_{ij}}+e^{\beta\Delta}}{e^{\beta\delta_{ij}}+e^{-\beta\Delta}}\right)\theta(\delta_{ij}-2\Delta)\right.\right.$$
$$\left.+2T\ln\frac{e^{\beta\Delta}+1}{e^{\beta\Delta}+e^{-\beta\delta_{ij}}}\theta(2\Delta-\delta_{ij})\right]$$
$$+\left[\frac{\varepsilon(\delta_{ij}-\varepsilon)+\Delta^2}{(\delta_{ij}-\varepsilon+\Delta)^{1/2}}\frac{2}{\delta_{ij}^{1/2}}K\left(\left(\frac{\delta_{ij}-2\Delta}{\delta_{ij}}\right)^{1/2}\right)\right.$$
$$\left.\left.-(\delta_{ij}-2\Delta)\right]e^{\beta\delta_{ij}}f_0(\varepsilon)f_0(\delta_{ij}-\varepsilon)\theta(\delta_{ij}-2\Delta)\right\}, \qquad \text{(A4)}$$

$$\beta_1=-\frac{\alpha\tau_{\text{эв}}\omega_0}{T}\{Xf_0(\Delta+\delta_{ij}+\omega_0)-Yf_0(\Delta+\delta_{ij})+\theta[1-f_0(\Delta)]$$
$$-R[1-f_0(\Delta)]-Sf_0(\delta_{ij}-\varepsilon_1)+Vf_0(\delta_{ij}-\varepsilon_2)-Uf_0(\varepsilon_3)+Wf_0(\varepsilon_2)\}, \qquad \text{(A5)}$$

$$\alpha_1=\beta_1+\frac{\alpha\tau_{\text{эв}}\omega_0}{T}\{X-Y+\theta-R-S+V-U+W\}, \qquad \text{(A6)}$$

where

$$X=\mathrm{ch}^{-2}\frac{\Delta+\omega_0}{2T}\varphi_1(\Delta+\omega_0,\omega_0)\frac{2}{(\delta_{ij}+\omega_0)^{1/2}}K\left(\left(\frac{\delta_{ij}}{\delta_{ij}+\omega_0}\right)^{1/2}\right),$$

$$Y=\mathrm{ch}^{-2}\frac{\Delta}{2T}\varphi_1(\Delta,-\omega_0)\frac{2}{\delta_{ij}^{1/2}}K\left(\left(\frac{\delta_{ij}-\omega_0}{\delta_{ij}}\right)^{1/2}\right),$$

$$\theta=\mathrm{ch}^{-2}\frac{\Delta+\delta_{ij}}{2T}\varphi_2(\Delta,-\omega_0)\frac{2}{(\delta_{ij}+\omega_0)^{1/2}}K\left(\left(\frac{\delta_{ij}}{\delta_{ij}+\omega_0}\right)^{1/2}\right),$$

$$R=\mathrm{ch}^{-2}\frac{\Delta+\delta_{ij}}{2T}\varphi_2(\Delta,\omega_0)\frac{2}{\delta_{ij}^{1/2}}K\left(\left(\frac{\omega_0}{\delta_{ij}}\right)^{1/2}\right),$$

$$S=\mathrm{ch}^{-2}\frac{\varepsilon_1}{2T}\xi_1(\varepsilon_1,\omega_0)\frac{1}{(\delta_{ij}-2\Delta)^{1/2}}$$
$$\times K\left(\left(\frac{\delta_{ij}-2\Delta-\omega_0}{\delta_{ij}-2\Delta}\right)^{1/2}\right)\theta(\delta_{ij}-2\Delta-\omega_0),$$

$$V=\mathrm{ch}^{-2}\frac{\varepsilon_2}{2T}\xi_1(\varepsilon_2,-\omega_0)\frac{1}{(\delta_{ij}-2\Delta+\omega_0)^{1/2}}$$
$$\times K\left(\left(\frac{\delta_{ij}-2\Delta}{\delta_{ij}-2\Delta+\omega_0}\right)^{1/2}\right)\theta(\delta_{ij}-2\Delta),$$

$$U=\mathrm{ch}^{-2}\frac{\delta_{ij}-\bar{\varepsilon}_3}{2T}\xi_2(\varepsilon_3,\omega_0)\frac{1}{(\delta_{ij}-2\Delta)^{1/2}}$$
$$\times K\left(\left(\frac{\delta_{ij}-2\Delta-\omega_0}{\delta_{ij}-2\Delta}\right)^{1/2}\right)\theta(\delta_{ij}-2\Delta-\omega_0),$$

$$W=\mathrm{ch}^{-2}\frac{\delta_{ij}-\varepsilon_2}{2T}\xi_2(\varepsilon_2,-\omega_0)\frac{1}{(\delta_{ij}-2\Delta+\omega_0)^{1/2}}$$
$$\times K\left(\left(\frac{\delta_{ij}-2\Delta}{\delta_{ij}-2\Delta+\omega_0}\right)^{1/2}\right)\theta(\delta_{ij}-2\Delta),$$

$$\varphi_1(\varepsilon,\omega_0)=\frac{[\varepsilon(\varepsilon+\delta_{ij})-\Delta^2][\varepsilon(\varepsilon-\omega_0)+\Delta^2]}{\varepsilon[(\varepsilon+\delta_{ij}+\Delta)(\varepsilon-\omega_0+\Delta)(\varepsilon+\Delta)]^{1/2}},$$

$$\varphi_2(\varepsilon,\omega_0)=\frac{[\varepsilon(\varepsilon+\delta_{ij})-\Delta^2][(\varepsilon+\delta_{ij})(\varepsilon+\delta_{ij}-\omega_0)+\Delta^2]}{(\varepsilon+\delta_{ij})[(\varepsilon+\delta_{ij}+\Delta)(\varepsilon+\delta_{ij}-\omega_0+\Delta)(\varepsilon+\Delta)]^{1/2}},$$

$$\xi_1(\varepsilon,\omega_0)=\frac{[\varepsilon(\delta_{ij}-\varepsilon)+\Delta^2][\varepsilon(\varepsilon-\omega_0)+\Delta^2]}{\varepsilon[(\delta_{ij}-\varepsilon+\Delta)(\varepsilon-\omega_0+\Delta)(\varepsilon+\Delta)]^{1/2}},$$

$$\xi_2(\varepsilon,\omega_0)=\xi_1(\delta_{ij}-\varepsilon,\omega_0),$$

$$\Delta<\bar{\varepsilon}<\delta_{ij}-\Delta,\quad \Delta+\omega_0<\bar{\varepsilon}_1<\delta_{ij}-\Delta,$$

$$\Delta<\bar{\varepsilon}_2<\delta_{ij}-\Delta,$$

$$\Delta<\bar{\varepsilon}_3<\delta_{ij}-\Delta-\omega_0,$$

and $K(z)$ is the complete elliptic integral of the first kind.[24]

[1)]In what follows we limit ourselves to investigating only two-level systems.

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