Electron-phonon interaction in a superconductor with Kondo scattering

A.G.Kozorezov, A. A. Golubov, J.K.Wigmore, D.Martin, P.Verhoeve, R.A. Hijmering

1 Department of Physics, Lancaster University, Lancaster, UK,
2 Department of Applied Physics, University of Twente,
P.O. Box 217, 7500 AE Enschede, The Netherlands,
3 Science Payloads and Advanced Concepts Office, SCI-A,
European Space Agency, ESTEC, Noordwijk, The Netherlands

(Dated: April 9, 2008)

In a superconductor with magnetic impurities, Kondo scattering results in the formation of localized states inside the superconducting gap. We show that inelastic electronic transitions involving quasiparticle scattering into and out of the localized states may result in significant changes in the non equilibrium properties of the superconductor. Using the model of Muller-Hartmann and Zittartz for the extreme dilute limit, and including both deformation potential and spin-lattice coupling we have calculated the rates of such inelastic transitions between continuum and discrete states, and shown that they may greatly modify quasiparticle interactions. The individual processes are: quasiparticle trapping into discrete states, enhanced recombination with localized quasiparticles, and pair breaking and detrapping of localized quasiparticles by sub-gap phonons. We find that all these processes give rise to clearly distinguishable temperature dependences of the kinetic parameters.

I. INTRODUCTION

The study of the effects of magnetic impurities in superconductors originated with the pioneering work by Abrikosov and Gor’kov\(^1\). Recent interest in the subject has been greatly stimulated by direct observation of the states bound to impurity atoms\(^2\), which has led to extensive experimental and theoretical work in both conventional and unconventional superconductors\(^3\). An important consequence of such intra gap bound states which has not previously been considered is their role in providing enhanced trapping and recombination at impurity atoms in analogy with deep levels in semiconductors\(^4\). Thus, quasiparticles initially in continuum states may undergo inelastic scattering with phonon emission and become localized in the vicinity of impurity atoms, which will act as recombination centers and provide rapid thermalization of a non equilibrium initial distribution. The formation of an intra gap band of impurity levels, possibly even overlapping the ground state, will modify the temperature dependence of thermalization. Finally, activation of localized quasiparticles into the continuum spectrum results in an anomalous temperature dependence of the observable parameters characterizing the non equilibrium state, such as quasiparticle lifetime\(^5\). Previously neither mechanisms of coupling nor transitions between the continuum and discrete states bound to impurities were discussed. In this paper we will show that electronic transitions between the continuum and bound states occur both due to deformation potential and spin-lattice interaction. As will be described later there is strong evidence that such a scenario has already been observed experimentally.

We will consider the dilute impurity limit \(c \ll 1\). Here \(c\) is the dimensionless impurity density in units of the condensate density \(\frac{2N(0)\Delta}{\Delta}\), where \(N(0)\) is the density of states at the Fermi level per spin in the normal state, and \(\Delta\) is the gap. The problem can be explicitly solved within the model originally developed by Müller-Hartmann and Zittartz\(^6\) for quantum spins in fully gapped superconductors. In this model the Hamiltonian of the system is taken in the form

\[
H = H_0 + H' \tag{1}
\]

where \(H_0\) is the Hamiltonian of an ideal superconductor and \(H'\) describes the interaction between impurity atoms and conduction electrons. The corresponding interaction potential has the form\(^6\),

\[
v(r) = \sum_i [u_1(r - R_i) + u_2(r - R_i) \sigma \cdot S] \tag{2}
\]

where \(R_i\) is the coordinate of the impurity atom \(S\) is its spin and \(\sigma_{x,y,z}\) are the spin Pauli matrices. Here the first term describes the spin independent part of the impurity scattering potential, and the second term the exchange interaction. To consider phonon assisted electronic transitions involving the bound states we must include also terms, describing the electron-phonon interaction through both the deformation potential and the spin-lattice interaction; these are derived by expanding the first and the second terms in expression \((2)\) respectively to include the displacement of the impurity atom from its equilibrium site. In the four-dimensional matrix formalism the full interaction Hamiltonian
describing phonon assisted electronic transitions has the form

\[ H_{\text{int}} = \frac{1}{2} \int d\mathbf{r} \psi^+(\mathbf{r}) V(\mathbf{r}) \psi(\mathbf{r}) \]  

(3)

where \( \psi^+(\mathbf{r}) \) and \( \psi(\mathbf{r}) \) are four-component operators and \( V(\mathbf{r}) \) is the 4\times4 matrix of the form \( \begin{pmatrix} \hat{v} & 0 \\ 0 & -\hat{v}^T \end{pmatrix} \), where \( \hat{v}(\mathbf{r}) = \sum_i Q_i \nabla (u_1 \sigma_0 + u_2 \sigma_2 \cdot \mathbf{S}), \hat{v}^T \) is the transposed matrix, and \( Q_i \) is the lattice displacement of the impurity due to vibrations.

In a superconductor described by the Hamiltonian \((1)\), for an impurity with antiferromagnetic exchange, bound states split off from the gap are formed. For the dilute limit the shift of the gap edge remains small, being proportional to the density of impurities. Therefore we may disregard all effects of modifications to the continuum quasiparticle spectrum relating to level shift or to broadening. Our objective is to determine the spatially averaged Green function for the continuum spectrum, with the interaction given by \((3)\). In the limit \( c \ll 1 \) spatial averaging can be carried separately for all elements of the Dyson equation. Indeed, the interaction loop itself will give a contribution proportional to the number of discrete states and hence to \( c \). Therefore, replacing the external Green functions by spatially averaged ones introduces inaccuracy in terms only of the order of \( c^2 \ll 1 \), so that the only function remaining to be averaged is that inside the loop. After spatial averaging, the Fourier transform to the momentum space, and analytical continuation from the imaginary to the real axis \( i\omega_n \to \omega + i\delta \), we may write an expression for the self energy

\[
\Sigma_{\text{ph}}(\mathbf{p}, \omega) = \begin{pmatrix} \Sigma_{1,ph}^{\sigma_0} & \Sigma_{2,ph}^{i\sigma_2} \\ -\Sigma_{2,ph}^{i\sigma_2} & \Sigma_{1,ph}^{\sigma_0} \end{pmatrix}
\]

(4)

Here \( G(\mathbf{p}', \mathbf{z}') \), \( \tilde{G}(\mathbf{p}', \mathbf{z}') \), \( F(\mathbf{p}', \mathbf{z}') \) and \( F^+(\mathbf{p}', \mathbf{z}') \) are spatially averaged electronic Green functions obtained within the model of Müller-Hartmann and Zittartz in the dilute limit. The coupling strength \( q_{\mathbf{q},j}^2(\mathbf{p}, \mathbf{p}') \) is introduced through

\[
\begin{align*}
|g_{\mathbf{q},j}(\mathbf{p}, \mathbf{p}')^2 &= |g_{\mathbf{q},j}^{(1)}(\mathbf{p}, \mathbf{p}')^2 \pm S(S+1)|g_{\mathbf{q},j}^{(2)}(\mathbf{p}, \mathbf{p}')^2 \\
|g_{\mathbf{q},j}^{1,2}(\mathbf{p}, \mathbf{p}')^2 &= \frac{\hbar}{2MN\omega_{\mathbf{q},j}}(\mathbf{p} - \mathbf{p}', \mathbf{e}_{\mathbf{q},j})^2 |u_{1,2}(\mathbf{p} - \mathbf{p}')^2 
\end{align*}
\]

(5)

We obtain for the rate of quasiparticle transitions from the state \( (\mathbf{p}, \epsilon) \) the following expression

\[
\Gamma(p) = -\frac{1}{2\epsilon Z_1(0)} [(\xi p + \epsilon) \text{Im} \Sigma_{1,ph} + (\xi p - \epsilon) \text{Im} \tilde{\Sigma}_{1,ph} - \Delta \text{Im} (\Sigma_{2,ph} + \Sigma_{2,ph}^+)]
\]

(6)

where \( Z_1(0) \) is the real part of the renormalization parameter. Introducing coupling constants analogous to the Eliashiberg constant

\[
\alpha_{1,2}^2(z) F(z) = \frac{\int_{S_F} d^2p |p| \int_{S_F} d^2p' |p'| \sum_{\mathbf{q},j} |g_{\mathbf{q},j}^{1,2}(\mathbf{p}, \mathbf{p}')^2 \delta(z - \omega_j(\mathbf{q}))}{\int_{S_F} d^2p |p|}
\]

(7)

we obtain

\[
\Gamma(\epsilon) = \frac{\pi}{Z_1(0)} \int_{-\infty}^{\infty} dz dz' F(z) \{\alpha_1^2(z) \text{Re} \left[ G_m(z') - \frac{\Delta}{\epsilon} F_m(z') \right] \}
\]
where denominators in (11). The simplified equations then become

\[
\{ \tanh \left( \frac{z'}{2\Delta} \right) + \coth \left( \frac{z}{2\Delta} \right) \} \delta(\epsilon - z - z') - \left\{ \tanh \left( \frac{z'}{2\Delta} \right) - \coth \left( \frac{z}{2\Delta} \right) \right\} \delta(\epsilon + z - z') \}
\]

(8)

Here we have introduced the new notations \( G_m(z) \) and \( F_m(z) \) for the spatially averaged Green functions. The exact expressions for the \( G_m(z) \) and \( F_m(z) \) have the form

\[
G_m(\epsilon) = \frac{\bar{\epsilon}(\epsilon)}{\sqrt{\epsilon^2(z) - \Delta^2(\epsilon)}}; \quad F_m(\epsilon) = \frac{\tilde{\Delta}(\epsilon)}{\sqrt{\epsilon^2(\epsilon) - \Delta^2(\epsilon)}}
\]

(9)

The energy \( \bar{\epsilon} \) and the order parameter \( \tilde{\Delta} \) satisfy the following equations:

\[
\bar{\epsilon} = \epsilon + \Delta \Sigma_1(\bar{\epsilon}, \tilde{\Delta}); \quad \tilde{\Delta} = \Delta + \Delta \Sigma_2(\bar{\epsilon}, \tilde{\Delta})
\]

(10)

where

\[
\Sigma_1(y, \Delta) = -c \sqrt{\frac{y^2}{2\Delta^2}} \frac{y(y - y_0)}{y^2 - y_0^2}
\]

\[
\Sigma_2(y, \Delta) = c \sqrt{\frac{y^2}{2\Delta^2}} \frac{y(y - y_0)}{y^2 - y_0^2}
\]

(11)

Here \( y = \epsilon/\Delta, y_0 = \epsilon_0/\Delta < 1 \) and \( \epsilon_0 \) is the discrete intra gap level. To find solutions to the main terms, we may use the simplified equations obtained from (11) by taking the renormalised parameters \( \bar{\epsilon} = \epsilon/\Delta \) and \( \tilde{y}_0 = \epsilon_0/\tilde{\Delta} \) in the denominators in (11). The simplified equations then become

\[
\bar{z} = z - c \frac{y}{|y|} \left( \frac{1}{y - y_0} - \frac{1}{y + y_0} \right)
\]

\[
\tilde{\Delta} = \Delta + \tilde{\epsilon} \frac{y}{|y|} \Delta \left( \frac{1}{y - y_0} - \frac{1}{y + y_0} \right)
\]

(12)

where \( \tilde{\epsilon} = c \sqrt{\frac{1 - y^2}{2y_0^2} (1 - y_0^2)} \). In order to solve the simplified equations we first note that from (11) \( \Sigma_2 = -y_0/\Sigma_1 \).

Above the gap edge both \( |\Sigma_1| \sim c \) and \( |\Sigma_2| \sim c; \) as pointed out above we ignore these corrections. Inside the gap the solution for the imaginary part \( \Sigma_1'' \) has the form

\[
\Sigma_1'' = \sqrt{\tilde{\epsilon} - \frac{1}{4}(|y| - y_0)^2} \Theta \left( \tilde{\epsilon} - \frac{1}{4}(|y| - y_0)^2 \right)
\]

(13)

Within the range \( \Sigma_1'' \neq 0 \), the real part of the self-energy \( \Sigma_1' \) is given by

\[
\Sigma_1' = \frac{1}{2}(y_0 - |y|) + \tilde{\epsilon} \frac{1}{4y_0}
\]

(14)

Outside this range, but still inside the gap, \( \Sigma_1' \) remains finite with a dependence on impurity concentration changing from \( \sqrt{\tilde{\epsilon}} \) at the edge of the \( \Sigma_1'' \neq 0 \) range to \( c \) away from it.

Finally, inside the gap we obtain

\[
\text{Re} G_m(y) = \frac{1 + y_0}{(1 - y_0)^{3/2}} \sqrt{\tilde{\epsilon} - \frac{1}{4}(|y| - y_0)^2} \Theta \left( \tilde{\epsilon} - \frac{1}{4}(|y| - y_0)^2 \right)
\]

\[
\text{Re} F_m(y) = \frac{y_0(1 + y_0)}{(1 - y_0)^{3/2}} \sqrt{\tilde{\epsilon} - \frac{1}{4}(|y| - y_0)^2} \Theta \left( \tilde{\epsilon} - \frac{1}{4}(|y| - y_0)^2 \right)
\]

(15)

These expressions describe the normalised density of bound states inside the gap in a superconductor with magnetic impurities. This distribution is sharp, with both its width and height being proportional to \( \sqrt{\tilde{\epsilon}} \). It is easy to confirm
that \( \int_{-1}^{1} dy \text{Re} G_m(y) = c \), corresponding to one quasiparticle bound state for each impurity atom. Inside the gap, from (15), we also obtain \( \text{Re} F_m(y) = y_0 \text{Re} G_m(y) \).

Using the Green functions given by (15) and (10) we may now analyze the different inelastic transitions. Firstly, a quasiparticle initially in the continuum state may become trapped. For the trapping rates we obtain

\[
\frac{1}{\tau_{\text{trap}}(\epsilon)} = \frac{2\Gamma(\omega)}{\hbar} = \int_0^{\Delta} \frac{d\epsilon'(\epsilon - \epsilon')}{\Delta^2}
\]

\[
[\text{Re} G_m(\epsilon') \left( \frac{1}{\tau_1} + \frac{1}{\tau_2} \right) - \frac{\Delta}{\epsilon} \text{Re} F_m(\epsilon') \left( \frac{1}{\tau_1} - \frac{1}{\tau_2} \right)] [n(\epsilon - \epsilon') + 1]
\]

\[
[1 - f(\epsilon')] = c \left[ \frac{1}{\tau_1} \left( \frac{1}{\epsilon - \epsilon_0} \right)^2 + \frac{1}{\tau_2} \frac{\epsilon^2 - \epsilon_0^2}{\Delta \epsilon} \right] [n(\epsilon - \epsilon_0) + 1]\]

where \( n(\epsilon) \) and \( f(\epsilon) \) are the phonon and quasiparticle distribution functions. The characteristic relaxation times for phonon assisted scattering on magnetic impurity are for deformation potential coupling, and \( T_c \) is the critical temperature. The top bars in this notation emphasize that these characteristic times are for phonon assisted scattering on a magnetic impurity. An order of magnitude estimate of the ratio \( \tau_0/\tau_{1,2} \) can be obtained by direct evaluation of factors \( \alpha^2_{1,2} \). Thus

\[
\frac{\tau_0}{\tau_1} \sim \left( \frac{u_1}{u_{ei}} \right)^2 \frac{\hbar v_s}{\Delta a_s} \frac{\tau_0}{\tau_2} \sim S(S + 1) \left( \frac{u_2}{u_{ei}} \right)^2 \frac{\hbar v_s}{\Delta a_s}
\]

where \( u_1, u_2 \) and \( u_{ei} \) are the characteristic values of electron-impurity, exchange interaction and electron-ion potentials respectively, \( v_s \) sound velocity and \( a_s \) is the radius of the bound state.

The recombination rate via a bound state calculated from (8) and (15) is given by

\[
\Gamma_{R,t}(\epsilon) = \int_0^{\Delta} \frac{d\epsilon'(\epsilon + \epsilon')}{\Delta^2}
\]

\[
[\text{Re} G_m(\epsilon') \left( \frac{1}{\tau_1} + \frac{1}{\tau_2} \right) + \frac{\Delta}{\epsilon} \text{Re} F_m(\epsilon') \left( \frac{1}{\tau_1} - \frac{1}{\tau_2} \right)] [n(\epsilon + \epsilon') + 1]f(\epsilon') = c \left[ \frac{1}{\tau_1} \left( \frac{1}{\epsilon + \epsilon_0} \right)^2 + \frac{1}{\tau_2} \frac{\epsilon^2 - \epsilon_0^2}{\Delta \epsilon} \right] [n(\epsilon + \epsilon_0) + 1]f(\epsilon_0)
\]

The expression can be written in a more familiar form by introducing the appropriate recombination coefficient \( R_t \) and density of trapped quasiparticles \( n_t \)

\[
\Gamma_{R,t}(\epsilon) = R_t n_t; \quad R_t = \frac{1}{2N(0)\Delta} \left[ 1 \left( \frac{\epsilon + \epsilon_0}{\tau_1} \right)^2 + \frac{1}{\tau_2} \frac{\epsilon^2 - \epsilon_0^2}{\Delta \epsilon} \right]
\]

which describes the maximum recombination rate in the absence of a phonon bottle-neck effect. Comparing the recombination coefficient on traps \( R_t \) with that in ideal superconductor \( R \) we obtain

\[
\frac{R_t}{R} \approx \left( \frac{u_1}{u_{ei}} \right)^2 + S(S + 1) \left( \frac{u_2}{u_{ei}} \right)^2 \frac{\hbar v_s}{\Delta a_s}
\]

Taking values for \((u_1/u_{ei})^2, (u_2/u_{ei})^2 \approx 1, v_s = 3\times10^9\text{ cm/s}, \Delta = 0.5\text{ meV}, S = 1\) and \( a_s = 1\text{ nm} \) we obtain \( R_t/R \geq 10 \). This ratio indicates the dominance of recombination at impurities due to the larger magnitude of the spin-lattice and deformation potential coupling constant with discrete levels originating from the appearance in (15) of a form-factor for phonon assisted impurity scattering, instead of the momentum conservation law. Similarly, comparing the maximum
recombination rates under quasi equilibrium conditions for the two different processes, we obtain

$$\frac{R \rho_{n,T}}{R \rho_T} = \frac{1}{4} \sqrt{\frac{2\Delta}{\pi T}} \exp \left( \frac{\Delta - \epsilon_0}{T} \right) \left[ \left( \frac{\Delta + \epsilon_0}{\Delta} \right)^2 \frac{\alpha_1^2}{\alpha^2} + \frac{\Delta^2 - \epsilon_0^2 \alpha_2^2}{\Delta^2} \right]$$

(22)

where $n_{T,T}$ and $n_T$ are thermal distributions of trapped and mobile quasiparticles. Hence, even for a small impurity density, recombination on the traps at low temperatures is a stronger process because of the presence of the exponential factor. The presence of this factor significantly accelerates recombination at low temperatures in superconductors containing concentrations of magnetic impurities which are below trace levels. Moreover, the possible formation of an intra gap band of bound states, and also of discrete bound states in the vicinity of the Fermi level, can significantly change the observed temperature dependence of recombination and thermalization rates. In some situations the rates in an impure superconductor may remain finite even at $T = 0$.

In spite of the stronger coupling constant the pair breaking by sub-gap phonons at small impurity density may be less efficient than for transitions in the continuum spectrum. In the latter case strong pair-breaking is known to slow down the recombination rate because of phonon bottle-necking. Thus less efficient phonon bottle-necking also enhances recombination at impurities. The pair breaking rate can be calculated using the Green functions calculated earlier to give

$$\Gamma_{pb}(\Omega) = \frac{2\tau_0}{\pi \tau_{ph}} \int_0^{\Omega - \Delta} \frac{d\epsilon}{\Delta} \left( \frac{1}{\tau_1} + \frac{1}{\tau_2} \right) \text{Re} G_m(\epsilon) \text{Re} G_m(\Omega - \epsilon)$$

$$+ \left( \frac{1}{\tau_1} - \frac{1}{\tau_2} \right) \text{Re} F_m(\epsilon) \text{Re} F_m(\Omega - \epsilon) = \frac{1}{\tau_{ph}} \eta(\Omega)$$

(23)

where $\tau_{ph}$ is the characteristic pair breaking time of an ideal superconductor and

$$\eta(\Omega) = \frac{1}{\pi^2 \epsilon} \left[ \frac{\tau_0}{\tau_1} + \left( 1 - 2\frac{\epsilon_0}{\Omega} \right) \frac{\tau_0}{\tau_2} \right] \frac{(1 + y_0)^{3/2}}{\sqrt{1 + \Omega/\Delta - y_0}}$$

$$\int_{\chi(\Omega,y_0)+2\sqrt{\epsilon}}^{\chi(\Omega,y_0)+2\sqrt{\epsilon}} dt \sqrt{4\epsilon - (\chi(\Omega,y_0) - t^2)^2}$$

(24)

where $\chi(\Omega,y_0) = \Omega/\Delta - 1 - y_0$ and $\Theta$ is the step function.

The rate of de-trapping from the localized state can be calculated without the need for direct evaluation of the broadening of the bound state due to spin-lattice interaction. Since we are interested in the de-trapping rate due to transitions into all available states, we may balance scattering into and out of the bound states at thermal equilibrium. The result is

$$\frac{1}{\tau_{de-trap}} = \frac{1}{\sqrt{2}} \int_{\frac{1}{\tau_1} - \frac{1}{\tau_2}}^\infty dz \exp \left( -z \frac{\Delta}{\Delta} \right) \left[ \frac{1}{\tau_1} z + \frac{1}{\tau_2} \left( 1 + \frac{\epsilon_0}{\Delta} \right) \right]$$

(25)

The formulas (10), (19), (23), and (27) may be compared with the well known results for an ideal superconductor.

Experimental data indicating the possible presence of such processes has appeared previously in several works. Thermalization which is several orders of magnitude faster than expected is routinely seen in a number of superconducting absorber materials used in low temperature single photon detectors. In our own earlier experiments on non stationary, non equilibrium quasiparticle distributions in superconducting tunnel junctions (STJs) we found that many detailed features of the experiments could only be successfully modelled on the assumption of local trapping states with well defined energy levels. However, in the absence of a microscopic model for the trap, the approach was purely phenomenological. All inelastic electronic transitions into and out of the traps were modelled with parameters determined from fitting to experimental data. In experiments on nominally pure Nb, Ta and Al films and various proximised bi-layer structures, we showed that the observed behaviour was consistent with the local traps being either macroscopic regions of suppressed gap as previously seen in low temperature SEM scans, or due to the presence of magnetic impurities.

Recently the anomalous temperature dependence of quasiparticle lifetimes in Ta and Al films similar to that observed in Nb was reported by R.Barends et. al detected through measurements of kinetic inductance and the observation of noise spectra. An important feature of the results obtained by this technique was the spatial homogeneity
of the response, indicating the intrinsic character of the traps and suggesting that they were associated with magnetic impurities distributed homogeneously throughout the superconductor. The new mechanisms for inelastic scattering of the electrons may also be important for electron decoherence in normal metals with Kondo impurities, currently the subject of great interest.\textsuperscript{11,12}

We acknowledge valuable discussions with T.M.Klapwijk, R.Barends and J.R.Gao.

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