Theory of Knight Shift and Spin-Lattice Relaxation Rates of Pu-115

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ABSTRACT

We calculated the Knight shift and spin-lattice relaxation rates of Pu-115 compounds assuming d-wave superconductivity in the presence of strong impurity scattering. We discuss the implications for recent measurements of the spin-lattice relaxation rate in the Pu-115 compound PuRhGa$_5$ by Sakai and coworkers [J. Phys. Soc. Jpn. 74, 1710 (2005)] and present a prediction for the corresponding Knight shift. In addition, we noticed a significant round-off of the spin-lattice relaxation rate $1/T_1$ just above the superconducting transition temperature that is not observed in the sister compound PuCoGa$_5$. It appears that in PuRhGa$_5$ superconductivity is mediated by spin fluctuations, too. This provides additional support to the scenario of superconducting pairing mediated by spin fluctuations in the Pu-115 compounds similar to the Ce-115 compounds and the high-temperature copper-oxide superconductors.

INTRODUCTION

The discovery of superconductivity in plutonium based systems such as PuCoGa$_5$[1] and PuRhGa$_5$[2] has stimulated the study of unconventional superconductivity and the pairing symmetry and mechanism in these materials. The symmetry of an unconventional superconductor is reduced compared to the symmetry of its normal state, thus resulting in many novel properties of the quasiparticle excitation spectrum. It is believed that the superconducting action in Pu-115 [PuMGa$_5$ with $M$=Co and Rh] derives itself from the unique character of the 5f electrons of plutonium [3]. The tetragonal crystal structure of PuMGa$_5$ is isostructural to that of the Ce-115 series [CeMIn$_5$].

The purpose of this study is to shed light on the superconducting pairing symmetry and possible pairing mechanism in the Pu-115 compounds. Very recently, Curro and coworkers [4] proposed, based on their measurements of the Knight shift and spin-lattice relaxation rates, that the Pu-115 compounds are bridging the superconducting and normal-state properties of the heavy-fermion Ce-115 and high-temperature copper-oxide superconductors. Therefore providing a means for tuning the interaction strength of antiferromagnetic spin fluctuations to intermediate values between both extreme limits [5].

The experimental techniques of nuclear magnetic resonance (NMR) and nuclear quadrupolar resonance (NQR) have been used successfully in the past to distinguish between the spin states of Cooper pairs (spin singlet vs. spin triplet pairing) and provide indirect information on the symmetry of the gap function – fully gapped vs. nodal lines or nodal points in the gap function on the Fermi surface. Both techniques probe directly the quasiparticle density of states and reveal indirect information about the pairing symmetry.

The standard explanation of power vs. exponential laws in the low-temperature behavior of thermodynamic and transport properties, for example, the spin-lattice relaxation rate $1/T_1$, comes from the difference of nodal and fully gapped excitation spectra in the superconducting state.
In clean nodal superconductors $1/T_1$ exhibits a nearly $T^3$ behavior far below the superconducting transition temperature $T_c$, while it is exponential for gapped superconductors. On the other side, deviations from this behavior, like the $T$-linear temperature dependence of $1/T_1$ at low temperatures, are explained by impurity effects in an unconventional superconductor (SC) with lines of nodes on the Fermi surface. Very recently, Sakai and coworkers [6] reported such a result for the spin-lattice relaxation rate of PuRhGa$_5$ in the superconducting phase. This behavior closely resembles the spin-lattice relaxation rates measured by Curro and coworkers for PuCoGa$_5$, which belongs to the same family of Pu-115 compounds, albeit with a $T_c$ nearly twice as high [4].

Here we give a detailed theoretical description of the spin-lattice relaxation rate and predict what should be observed for the Knight shift if measured on the same sample. Our self-consistent treatment of impurity scattering in the superconducting state goes beyond the two-fluid approach used by Sakai et al. [6], which was used to explain the large residual density of states in PuRhGa$_5$. Simultaneous measurements of spin-lattice relaxation rate and Knight shift will place stringent constraints on the symmetry and magnitude of the superconducting gap function, as well as on the concentration and scattering strength of impurities at low temperatures.

**THEORY**

The effect of impurity scattering is included within the self-consistent $T$-matrix approximation [7,8,9,10,11]. For the case of particle-hole symmetry of the quasiparticle excitation spectrum the Nambu component $T_3$ of the $T$ matrix vanishes, and for a d-wave order parameter (OP) with isotropic scattering $T_1 = 0$ (also without loss of generality we can choose $T_2 = 0$ by general $U(1)$ gauge symmetry), where $T_i$ is the $i$th component of the $2 \times 2$ Nambu matrix expanded in Pauli matrices. Then we need to calculate only $T_0(\omega)$. The impurity self-energy is given by $\Sigma_0 = \Gamma T_0$, where $\Gamma = n_i/\pi N_0$. Here $N_0$ is the normal density of states (DOS) at the Fermi surface (FS), $n_i$ is the impurity concentration; $T_0(\omega_n) = \frac{g_0(\omega_n)}{|\omega_n^2 - g_0(\omega_n)|}$, where $g_0(\omega_n) = \frac{1}{\pi N_0} \sum_k \frac{\tilde{\omega}_n}{\tilde{\omega}_n^2 + \xi^2 + \Delta^2(k)}$. The impurity renormalized Matsubara frequency is defined by $\tilde{\omega}_n = \omega_n + \Sigma_0$, with $\omega_n = \pi T(2n + 1)$, and the scattering strength parameter $c$ is related to the s-wave phase shift $\delta_0$ by $c = \cot(\delta_0)$. Using this self-energy $\Sigma_0$ the following gap equation is solved self-consistently,

$$\Delta(\phi) = -N_0 g(\phi) \int \frac{d\phi'}{2\pi} V(\phi - \phi') T \sum_{\omega_n} \int_{-\omega_c}^{\omega_c} d\epsilon \frac{\Delta(\phi')}{\omega_n^2 + \epsilon^2 + \Delta^2(\phi')} , \tag{1}$$

where $V(\phi - \phi')$ is the angular parametrization of the pairing interaction, and $\omega_c$ is a typical cutoff energy. We assume the canonical d-wave gap function of the form $\Delta(k) = \Delta_0 (\cos k_x - \cos k_y)$ or $\Delta(\phi) = \Delta_0 \cos(2\phi)$ for a cylindrical Fermi surface. The pairing potential $V(\phi - \phi')$ induces a gap with d-wave symmetry. Although its microscopic origin is not the issue of this paper, we believe it originates from antiferromagnetic (AFM) spin fluctuations. The static limit of the spin susceptibility of the AFM fluctuations, $\chi(q, \omega = 0) \sim \frac{1}{(q-Q)^2 + \xi^2}$, is parameterized near the AFM wave vector $Q$ as [12]

$$V(\phi - \phi') = V_d(b) \frac{b^2}{(\phi - \phi' \pm \pi/2)^2 + b^2} , \tag{2}$$

where the parameter $b$ is inverse proportional to the AFM correlation length $\xi$, normalized by the cylindrical FS ($\xi \sim a \pi/b$; $a$ is the lattice parameter). For all calculations in this paper, we chose $b = 0.5$ which is not a sensitive parameter for our results unless $\xi$ is very large ($b < 0.1$) [12], i.e., within the range of $0.1 < b < 1$ our results show little variations and are qualitatively the same.
With the gap function $\Delta(\phi)$ and $T_0(\omega)$ obtained from Eq. 11 ($T_0(\omega)$ is analytically continued from $T_0(\omega_\nu)$ by Padé approximant method), we calculate the $1/T_1$ nuclear spin-lattice relaxation rate [4] [7] [8] [13]

$$\frac{1}{T_1T} \sim -\int_0^\infty \frac{\partial f_F(\omega)}{\partial \omega} \left[ \left\langle Re \frac{\tilde{\omega}}{\sqrt{\tilde{\omega}^2 - \Delta^2(\phi)}} \right\rangle^2_\phi + \left\langle Re \frac{\Delta(\phi)}{\sqrt{\tilde{\omega}^2 - \Delta^2(\phi)}} \right\rangle^2_\phi \right],$$

and the superconducting spin susceptibility $\chi_s$

$$\frac{\chi_s}{T} \sim -\int_0^\infty \frac{\partial f_F(\omega)}{\partial \omega} \left\langle Re \frac{\tilde{\omega}}{\sqrt{\tilde{\omega}^2 - \Delta^2(\phi)}} \right\rangle_\phi,$$

where $f_F(\omega)$ is the Fermi-Dirac function, the impurity renormalized quasiparticle energy $\tilde{\omega} = \omega + \Sigma(\omega)$, and $\langle ... \rangle_\phi$ means the angular average over the FS. The first term in the bracket of Eq. 11 is $N^2(\omega)$. The second term vanishes in our calculations because of the symmetry of the OP. To calculate $1/T_1T$ using Eq. 11, or $\chi_s$ using Eq. 11, we need the full temperature dependent gap function $\Delta(\phi, T)$ and $T_c$. Our gap equation Eq. 11 is the BCS gap equation, therefore it gives the BCS temperature behavior for $\Delta(\phi, T)$ and $\Delta_0 = 2.14 k_BT_c$ for the standard weak-coupling d-wave SC. In order to account for strong-coupling effects we use the phenomenological formula $\Delta(\phi, T) = \Delta(\phi, T = 0) \Xi(T)$ with $\Xi(T) = \tan(\beta \sqrt{T_c/T} - 1)$, and parameters $\beta$ and $\Delta_0/T_c$. Then we only need to calculate $\Delta(\phi, 0)$ at zero temperature. The temperature dependence of $\Sigma(\omega, T)$ ($= \Gamma T_0(\omega, T)$) is similarly extrapolated: $T_0(\omega, T) = T_0(\omega, T = 0) \Xi(T) + T_{\text{normal}}(1 - \Xi(T))$, where $T_{\text{normal}} = \Gamma/(\epsilon^2 + 1)$ is the normal state $T_0$. In our numerical calculations we chose $\beta = 1.74$, because our final results are not very sensitive with respect to this parameter, while the ratio $\Delta_0/k_BT_c$ is an important parameter to simulate strong-coupling effects. The larger this ratio is, the more important are strong-coupling effects.

RESULTS AND DISCUSSIONS

In figures 11 and 2 the spin-lattice relaxation rate of PuRhGa$_5$ by Sakai et al. [6] is shown, where $1/T_1$ is normalized to $1/T_1 = 10$ at $T = T_c$ for ease of comparison. The insets show the corresponding normalized quasiparticle DOS for varying scattering rates $\Gamma$. With our earlier described choice of parameters the impurity scattering rate $\Gamma/\Delta_0 = 0.032$ is enough to completely fill the low energy gap with impurity states and $N(\omega = 0)$ reaches more than 25% of the normal-state DOS $N_0$

In Fig. 11 we obtain a better fit to the experimental data (symbols) assuming a slightly lower superconducting transition temperature $T_c = 7.6$ K than their reported value of $T_c = 8.5$ K. This could indicate the presence of a pseudogap similar to the high-temperature superconductor YBa$_2$Cu$_3$O$_{7-\delta}$, where $1/T_1$ is suppressed just above $T_c$.

For the temperature dependence of the gap, we chose the parameters $\beta = 1.74$ and $2\Delta_0/k_BT_c = 5$ for the d-wave gap to account for the strong-coupling effects of superconductivity as explained before. As expected from the DOS results, due to the impurity induced residual states, $1/T_1$ displays the linear-$T$ dependence at low temperatures and the region of $T$-linear behavior increases with impurity concentration. For a higher value of $\Gamma/\Delta_0 = 0.064$, this $T$-linear region extends up to $0.35 T_c$. At temperatures near $T_c$ the coherence peak is almost invisible because of the sign-changing gap function, i.e., vanishing of the second term in Eq. 11. Below $T_c$ it shows a nearly $T^3$ behavior due to the lines of nodes in the gap until it goes through a gradual crossover
FIG. 1: The NQR spin-lattice relaxation rate plotted versus temperature normalized by $T_c$. Calculations are for $2\Delta_0 = 5k_B T_c$ and three values of the impurity scattering rate $\Gamma$ for unitary scattering. Inset: The normalized quasiparticle DOS for corresponding values of $\Gamma/\Delta_0 = 0, 0.032, 0.064$.

FIG. 2: The NQR spin-lattice relaxation rate plotted versus temperature normalized by $T_c$. Calculations are for $2\Delta_0 = 8k_B T_c$ and three values of the impurity scattering rate $\Gamma$ for unitary scattering. Inset: The normalized quasiparticle DOS for corresponding values of $\Gamma/\Delta_0 = 0, 0.032, 0.064$. 
FIG. 3: The calculated spin susceptibility $\chi_S$ of a d-wave SC normalized by its normal state value $\chi_N$ for gap values $2\Delta_0 = 5k_BT_c$ (solid lines) and $8k_BT_c$ (dotted lines), and impurity scattering $\Gamma/\Delta_0 = 0$ and 0.032.

region and finally to the $T$-linear region. The comparison with the experimental data by Sakai et al. [6] on PuRhGa$_5$ is in good agreement with unitary scattering, a gap value $2\Delta_0 = 5k_BT_c$, and a scattering rate close to $\Gamma/\Delta_0 = 0.032$. Based on this value, we estimate the superconducting transition temperature of the pristine sample to be $T_0 = T_c + \pi/4 \Gamma \approx 8.1$ K or 9.0 K, depending on the value of $T_c = 7.6$ K or 8.5 K.

In Fig. 2, the normalized $1/T_1$ is plotted for an enhanced strong-coupling d-wave gap value $2\Delta_0 = 8k_BT_c$, as was recently found for PuCoGa$_5$ [4]. Due to the larger gap value, the calculated $1/T_1$ is always less than the measured spin-lattice relaxation rate. Hence we find a poorer fit to the experimental data for this choice of the strong-coupling gap.

Fig. 3 shows the prediction for the spin susceptibility, $\chi_S$, or its corresponding NMR Knight shift, $K = K_0 + A\chi_S$, where $K_0$ and $A$ are constants for most materials. $\chi_S$ is calculated for the same d-wave gap values as was used for the spin-lattice relaxation rates in figures 1 and 2. Again a modest impurity scattering rate of $\Gamma/\Delta_0 = 0.032$ results in a large residual susceptibility at zero temperature, equivalent to roughly 25% of the normal state DOS or spin susceptibility $\chi_N$. The quantitative difference in the spin susceptibility between gap values $2\Delta_0 = 5k_BT_c$ and $8k_BT_c$ should be easily discernible in measurements of the Knight shift.

**CONCLUSIONS**

The NQR spin-lattice relaxation rate $1/T_1$ in PuRhGa$_5$ is consistent with a strong-coupling d-wave gap function and unitary impurity scattering similar to the observed behavior in its sister compound PuCoGa$_5$. Inspite of many similarities between PuCoGa$_5$ and PuRhGa$_5$, we also find
marked differences: (1) The maximum superconducting gap value of PuRhGa$_5$ is smaller than for PuCoGa$_5$, i.e., it is $2\Delta_0/k_B T_c = 5$ for PuRhGa$_5$ versus 8 for PuCoGa$_5$. This suggests that the fluctuations of the pairing bosons are weaker for PuRhGa$_5$, possibly hinting at a progressive trend for the strength of the spin-fluctuations in this class of materials. (2) Although the PuRhGa$_5$ sample was of similar age as PuCoGa$_5$ when measured, it had a three times larger relative scattering rate $\Gamma/\Delta_0 = 0.032$ compared to PuCoGa$_5$ with $\Gamma/\Delta_0 = 0.01$, which could be due to variations of the isotope mix of plutonium between both samples. (3) $1/T_1$ exhibits a rounded behavior between $T = 7.6$ K and 9 K resembling the pseudogap phenomenon in YBa$_2$Cu$_3$O$_{7-\delta}$, while no such rounding is observed for PuCoGa$_5$. This certainly needs clarifications by further studies of $1/T_1$ in the normal state, which will be reported in a separate paper.

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