Broken time-reversal symmetry probed by muon spin relaxation in the caged type superconductor Lu$_5$Rh$_6$Sn$_{18}$; SUPPLEMENTAL MATERIAL

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(Dated: November 25, 2014)

Here we expound in detail our group-theoretical arguments about the symmetry of the superconducting order parameter. We also present the associated nodal structure of the quasiparticle spectrum. The analysis presented here applies to any superconductor with $D_{4h}$ point group symmetry, broken time-reversal symmetry and strong spin-orbit coupling. Besides Lu$_5$Rh$_6$Sn$_{18}$, the ruthenate superconductor Sr$_2$RuO$_4$ has been recently argued to fall within this category [1]. We note, however, that our present analysis does allow for singlet-triplet mixing as put forward in reference [1].

Barring an independent magnetic transition whose critical temperature is fine-tuned to coincide with the superconducting critical temperature, the sudden increase in the muon spin relaxation at $T_c$ suggests that the superconducting state breaks time-reversal symmetry. Assuming that the superconductivity is static and does not break the translational symmetry of the lattice, it can be characterised by a momentum-dependent pairing potential $\Delta_{\alpha,\beta}(k)$. Here $\alpha, \beta = \uparrow$ or $\downarrow$ are the spin indices of the two electrons in a Cooper pair and $\hbar k$ is the momentum of one of the electrons with respect to the centre of mass of the pair. Standard symmetry analysis [2, 3] yields $\Delta_{\alpha,\beta}(k) = \sum_{n=1}^{D} \Gamma_n^{\alpha,\beta}(k)$ just below $T_c$, where the functions $\Gamma_n^{\alpha,\beta}$ form a basis set of one of the irreducible representations of the point group of the crystal, of dimension $D$. The form of the coefficients $\Delta_n$ is obtained by minimizing generic free energies of the appropriate symmetry. The superconducting state breaks time-reversal symmetry if $D > 1$ and two or more coefficients have different complex phases [2]. The quasiparticle spectrum is then given by the diagonalisation of the Bogoliubov-de Gennes Hamiltonian

$$H_{\text{BdG}} = \begin{pmatrix}
\epsilon(k) - \mu & \Delta_{\uparrow\uparrow}(k) & \Delta_{\uparrow\downarrow}(k) \\
0 & \epsilon(k) - \mu & \Delta_{\downarrow\uparrow}(k) \\
\Delta_{\uparrow\downarrow}(k) & -\epsilon(k) + \mu & 0
\end{pmatrix},$$

where $\epsilon(k)$ is the single-electron dispersion relation, measured from the chemical potential. Note that in non-centrosymmetric systems (not considered here) the off-diagonal elements in the single-electron dispersion relation would be essential.

For the space group $I4_1/acd$ the relevant point group is tetragonal $D_{4h}$ [4] which has been thoroughly examined in the context of cuprate superconductivity [2, 3]. Let us first consider the case of singlet pairing. Quite generally, $\Delta(k) = \Delta_0(k)i\tilde{\sigma}_y$ (where $\tilde{\sigma}_y$ is the second Pauli matrices). Following Ref. [2], for the point group of interest, there are 7 possible instabilities, only one of which (corresponding to the $1E_g$ irrep) breaks time-reversal symmetry. The gap function in this case has the form [2]

$$\Delta_0(k) = (X + iY)Z$$

where $X, Y, Z$ are three real functions of $k$ that transform as $k_x, k_y$ and $k_z$, respectively, under the point group symmetry operations. Fig. 1 (a) depicts the size, at the Fermi surface, of the corresponding gap in the quasiparticle energy spectrum, $\propto \Delta_0(k)$, obtained by assuming an isotropic single-electron dispersion relation, $\epsilon(k) = \hbar^2|k|^2/2m^*$ (yielding a spherical Fermi surface) and taking the simplest forms for the functions $X, Y, Z$, namely $k_x, k_y$ and $k_z$, respectively. The gap is given as follows

$$\Delta(k) \propto |k_z| \sqrt{k_x^2 + k_y^2}$$

and so the low energy excitations would be dominated by a line node at the equator ($k_z = 0$), leading to a specific heat proportional to $T^2$ at low temperatures $T \ll T_c$ (the only exception being if the Fermi surface does not traverse the $k_z = 0$ plane, which is unlikely). In addition,
FIG. 1: Four possible forms of the gap in the quasi-particle energy spectrum, plotted on a spherical Fermi surface. (a) Singlet gap function [Eq. (2)] showing a line node on the equator and two linear point nodes at the “North” (N) and “South” (S) poles of the Fermi surface.

In the case of triplet pairing, i.e. \( \hat{\Delta}(\mathbf{k}) = i [\mathbf{d}(\mathbf{k}) \cdot \hat{\sigma}] \hat{\sigma}_y \), the double group combining the point group of the crystal with spin rotations has to be considered. Still following [2], for \( D_{4h} \) we find once more that only one of 7 possible instabilities (corresponding to the \( E_u(c) \) irrep of the double group) breaks time-reversal symmetry. The \( \mathbf{d} \)-vector in this case is given by [2]

\[
\mathbf{d}(\mathbf{k}) = (AZ,iAZ,B(X+iY))
\]

where the real coefficients \( A \) and \( B \) depend on details of the band structure and effective electron-electron interactions. This gap function corresponds to non-unitary triplet pairing as \( \mathbf{d}^* \times \mathbf{d} \neq 0 \).

The gap for this triplet case, with the same simplifying assumptions used above, is depicted in Fig. 1 (b-d). Its formula is

\[
\Delta(\mathbf{k}) \propto \left| A|k_z| - \sqrt{A^2 k_z^2 + B^2 (k_x^2 + k_y^2)} \right|
\]

Interestingly, the spectrum also features two point nodes at \( X = Y = 0 \), but in this case the point nodes are “shallow” using the terminology of [5]. Indeed this instability is analogous to the \( E_{2u} \) instability proposed for \( \text{UPt}_3 \) which also features a shallow node [6]. These nodes become ordinary linear nodes when \( B \gg A \), while they expand to cover the whole Fermi surface in the opposite limit, \( B \ll A \) (gapless superconductivity). Thus we expect the power-law exponent \( n \) characterising the low-temperature behaviour of the specific heat, \( C \sim T^n \), to be 1, 2, and 3 for the cases represented in panels (b), (c) and (d), respectively.

Non-unitary triplet pairing leads to the breaking of the two-fold degeneracy between the spin-up and spin-down parts of the quasi-particle spectrum. In this case we expect the superconducting instability to be accompanied by a bulk magnetisation that grows linearly with \( T_c - T \) for \( T \lesssim T_c \) and which acts as a sub-dominant order parameter [7, 8]. The linear increase of the muon spin relaxation rate \( \lambda \) that we observe experimentally (see Fig. 4 of the main text) also increases linearly below \( T_c \), which suggests that \( \lambda \) is simply proportional to this (very small [8]) bulk magnetisation.

We emphasise that the power laws mentioned above are only expected to be realised in the limit of very low temperatures (\( T \ll T_c \)). Moreover, if the topology of the Fermi surface departs significantly from a sphere the spectrum may become fully-gapped i.e. \( C \sim e^{-\Delta/T} \) for \( T \ll T_c \). Specifically, the triplet pairing potential may lead to a fully-gapped spectrum if the Fermi surface is open at the top and the bottom, so that it never cuts the N and S poles (e.g. a warped cylinder). In contrast the singlet order parameter would lead to line nodes with \( C \sim T^2 \) at low temperatures always. Finally, we point
out that a broader range of possibilities emerge if spin-orbit coupling happens to be weak enough to be neglected [2]. In that case two or more non TRS-breaking instabilities may merge to give new TRS-breaking ones [9].
