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Unveiling odd-frequency pairing around a magnetic impurity in a superconductor

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We study the unconventional superconducting correlations caused by a single isolated magnetic impurity in a conventional s-wave superconductor. Due to the local breaking of time-reversal symmetry, the impurity induces unconventional superconductivity which is even in both space and spin variables but odd under time inversion. We derive an exact proportionality relation between the even-frequency component of the local electron density of states and the imaginary part of the odd-frequency local pairing function. Finally, we apply this relation to scanning tunneling microscopy spectra taken on top of magnetic impurities immersed in a Pb/Si(111) monolayer and explicitly extract the odd-frequency superconducting pairing function.

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Introduction. Due to Fermi-Dirac statistics, the two-electron pairing correlation function at different times t_1 and t_2 has to be anti-symmetric under exchange of the two electrons or equivalently under the exchange of all their labels. These include time, spin, position and possibly other orbital degrees of freedom. In the conventional s-wave superconductor, the pairing function corresponds to an equal time, s-wave and spin-singlet pairing (assuming a single band) while the coveted p-wave superconductor corresponds to an equal time, p-wave, spin-triplet pairing function [1]. In the former case the sign change of the pair amplitude is provided by the spin variable, while in the latter by the space one. However there is a possibility that the sign may change under exchange of the two different time coordinates $t_1 \neq t_2$. More than four decades ago, Berezinskii proposed this possibility, the odd-frequency (odd- ω) pairing (thus odd under time exchange) in the s-wave triplet pairing of He³ [2] (see [3, 4] for recent reviews of this long lasting field). It was subsequently considered that odd- ω pairing can also be intrinsically generated in superconductors [5–7], or in heavy fermions compounds described by a Kondo lattice model [8–10].

The field boomed when Bergeret *et al.* realized that odd- ω pairing should appear in heterostructures made of a conventional s-wave superconductor and a ferromagnet [11, 12]. Such a platform has the key advantage of realizing odd- ω pairing in a controllable fashion with well understood materials. Conversely to the previous studies, the odd- ω pairing in these hybrid structures is the result of a proximity effect where the ferromagnet induces a spin-singlet to spin-triplet conversion of Cooper pairs. Such conversion actually allows Cooper pairs to propagate robustly far away in the ferromagnet which has opened the exciting possibility to achieve spintronics with superconductors [13, 14]. Subsequent studies demonstrated that odd-frequency pairing in fact appears in a wide variety of physical systems as a result of symmetry breaking. For example, odd- ω pairing can be realized

in non-magnetic junctions due to spatial parity breaking at the interface [15–17], which allows the conversion from s-wave to p-wave orbital symmetry. According to these predictions, odd- ω pairing should be rather ubiquitous in hybrid systems. However, there is not yet any clear and direct experimental evidence of odd- ω superconductivity though spectroscopic signatures in the density of states were reported in Nb superconducting films proximity coupled to epitaxial Ho [18].

Here we prove the existence of odd- ω pairing in the simplest hybrid system: a single magnetic impurity immersed in a conventional s-wave, spin singlet, even- ω superconductor. Our calculations show that on the magnetic impurity site a s-wave (local), spin triplet and odd- ω superconducting component arises from the breaking of the rotational symmetry. We establish an exact proportionality relation between the even- ω component of the local-impurity electron density of states (LDOS) and the imaginary part of the odd- ω superconducting function and provide expressions for the proportionality coefficients, which only depend on the parameters characterizing the magnetic impurity. We apply these results to account for the local density of states measured with scanning tunnelling spectroscopy (STS) on top of magnetic impurities immersed in a superconducting monolayer of Pb/Si(111). This provides the evidence of the presence of the odd- ω pairing component. Moreover, we are able to extract and explicitly display the superconducting odd- ω pairing function.

Local pairing functions. Due to the fermionic anti-commutation relations, the retarded and advanced superconducting functions $F_{\alpha,\beta}^R = -i\theta(t-t')\langle\{c_\alpha(t), c_\beta(t')\}\rangle$, $F_{\alpha,\beta}^A = i\theta(t'-t)\langle\{c_\alpha(t), c_\beta(t')\}\rangle$ are related, $F_{\alpha,\beta}^R(t, t') = -F_{\beta,\alpha}^A(t', t)$, or in frequency space $F_{\alpha,\beta}^R(\omega) = -F_{\beta,\alpha}^A(-\omega)$, under particle exchange. $\alpha, \beta = (\vec{r}, \uparrow, \downarrow \dots)$ are a priori any relevant set of quantum numbers, depending on the system. As here we study the local impurity ($\vec{r} = 0$), only spin variables are considered $\alpha, \beta = (\uparrow, \downarrow)$. Choosing the gauge where the order-parameter of the bare BCS

superconductor is real, the advanced and retarded pairing functions are related by complex conjugation. Thus, there are only two possible ways for the local retarded pairing functions to satisfy these relations,

$$\begin{aligned} 1. F_{\uparrow,\downarrow}^R(\omega) &= F_{\uparrow,\downarrow}^{R*}(-\omega) & \text{Even } \omega ; \text{ spin singlet,} \\ 2. F_{\uparrow,\downarrow}^R(\omega) &= -F_{\uparrow,\downarrow}^{R*}(-\omega) & \text{Odd } \omega ; \text{ spin triplet.} \end{aligned} \quad (1)$$

It is therefore convenient to decompose $F_{\uparrow,\downarrow}^R(\omega)$ in even- ω (spin-singlet) and an odd- ω (spin-triplet) components:

$$F_{\text{even}}^R(\omega) = \frac{1}{2}[F_{\uparrow,\downarrow}^R(\omega) + F_{\uparrow,\downarrow}^{R*}(-\omega)], \quad (3)$$

$$F_{\text{odd}}^R(\omega) = \frac{1}{2}[F_{\uparrow,\downarrow}^R(\omega) - F_{\uparrow,\downarrow}^{R*}(-\omega)]. \quad (4)$$

This implies that $\Re F_{\text{even}}^R(\omega)$, $\Im F_{\text{odd}}^R(\omega)$ are even functions while $\Im F_{\text{even}}^R(\omega)$, $\Re F_{\text{odd}}^R(\omega)$ are odd functions of frequency (here \Re, \Im correspond to the real and imaginary part respectively). A $F_{\text{odd}}^R(\omega)$ component in the total superconducting function is therefore the fingerprint of odd- ω superconductivity. The difficulty in proving the existence of odd- ω superconductivity relies then in extracting the superconducting function from spectral quantities. Our next goal is to show that $F_{\text{odd}}^R(\omega)$ can be indeed related and extracted from LDOS measured obtained with STS on a magnetic impurity site.

Model Hamiltonian and Dyson equation. Here we consider a single impurity in an s-wave superconductor. We assume the superconducting substrate to be infinite and homogeneous. This hybrid system can be described by a Bogoliubov-de Gennes (BdG) Hamiltonian which reads

$$\begin{aligned} \mathcal{H} &= \sum_{\mathbf{k}} \Psi^\dagger(\mathbf{k}) \begin{pmatrix} \varepsilon(\mathbf{k}) & \Delta \\ \Delta & -\varepsilon(\mathbf{k}) \end{pmatrix} \Psi(\mathbf{k}) \\ &+ \sum_{\mathbf{k}, \mathbf{k}'} \Psi^\dagger(\mathbf{k}) \begin{pmatrix} V - J & 0 \\ 0 & -(V + J) \end{pmatrix} \Psi(\mathbf{k}') \end{aligned} \quad (5)$$

where $\Psi^T(\mathbf{k}) = (c_\uparrow(\mathbf{k}), c_\downarrow^\dagger(\mathbf{k}))$ is a 2-component Nambu spinor. The superconductor is characterized by the metallic dispersion relation $\varepsilon(\mathbf{k})$ and a real pairing potential Δ . The magnetic impurity is modeled as a classical exchange field of strength J and a potential scattering V . We neglect any momentum dependence of these local couplings as they do not play any role here. This corresponds to the standard Yu-Shiba-Rusinov (YSR) model [19–21].

We use the Nambu-Gorkov Green function to completely describe the local one-particle electronic properties of the system

$$\hat{G}^R(t, t') = \begin{bmatrix} G_{\uparrow}^R(t - t') & F_{\uparrow,\downarrow}^R(t - t') \\ -F_{\downarrow,\uparrow}^R(t - t')^* & -G_{\downarrow}^R(t - t')^* \end{bmatrix}, \quad (6)$$

where $G_{\uparrow}^R(t - t') = -i\theta(t - t')\langle\{c_\uparrow(t), c_\uparrow^\dagger(t')\}\rangle$, and $F_{\uparrow,\downarrow}^R(t - t') = -i\theta(t - t')\langle\{c_\uparrow(t), c_\downarrow(t')\}\rangle$. The symbol $\langle\ldots\rangle$ is a short-hand notation denoting thermal average with respect to the full Hamiltonian.

The full Nambu-Gorkov local Green function in Fourier space at the position of the impurity can be computed using the Dyson equation

$$\hat{G}^R(\omega) = \hat{g}^R(\omega) + \hat{g}^R(\omega) \hat{\Sigma} \hat{G}^R(\omega) \quad (7)$$

where $\hat{g}^R(\omega)$ is the local Green function of the bulk superconductor in the absence of the magnetic impurity (*i.e.* the bare one) and

$$\hat{\Sigma} = \begin{bmatrix} V - J - i\Gamma & 0 \\ 0 & -(V + J) - i\Gamma \end{bmatrix}. \quad (8)$$

is the local self-energy in Nambu-space at the position of the impurity. Note that we also include a phenomenological imaginary part to this self-energy. The Dyson equation is easily solved as

$$\hat{G}^R(\omega) = [[\hat{g}^R(\omega)]^{-1} - \hat{\Sigma}]^{-1}. \quad (9)$$

Note the only assumptions we made at this point are that there is s-wave pairing and that the spin is locally a good quantum number. For $|\omega| < \Delta$ inside the superconducting gap, the bare green function is real in our gauge choice. Therefore the imaginary part of $\hat{G}^R(\omega)$ are Dirac distributions located at the poles of $1/\text{Det}[1 - \hat{g}^R(\omega)\hat{\Sigma}]$. These are the well-known YSR in-gap spin-polarized bound states [19–22]. Let us assume that these poles are well-separated in energy. Denoting by E_0 the bound state energy of one such pole, one can obtain an approximate expression of the LDOS as a function of J, V and of the expressions of the bare Green's functions taken at $\omega = E_0$.

Local density of states and odd- ω pairing. Due to recent enormous progress in the energy and spatial resolution of STS, YSR states are now very well characterized experimentally (see [23] for a recent review). It is worth stressing that the physics of chains of magnetic atoms on a superconducting substrate has attracted a considerable attention in the past years [24–44] as recent experiments on such systems have revealed the existence of zero bias peaks spatially localized on the ends of such chains which have been interpreted as signatures of Majorana bound states [45–50]. We shall now focus on the YSR states, and show that they can be employed to disentangle the even and odd frequency parts of the superconducting function.

The electronic LDOS measured by the differential conductance in STS is defined by $\rho(\omega) = -\frac{1}{\pi} \Im \left\{ \hat{G}_{11}^R(\omega) + \hat{G}_{22}^R(-\omega) \right\}$. After some algebra, the LDOS can be expressed as a linear combination of odd/even-frequency pairings as

$$\rho(\omega) \approx C_e(E_0) \times \Im F_{\text{odd}}^R(\omega) + C_o(E_0) \times \Im F_{\text{even}}^R(\omega). \quad (10)$$

where

$$C_e(E_0) = \frac{2JA(E_0) - g_{\uparrow}^R(E_0) + g_{\downarrow}^R(-E_0)}{\pi f_{\uparrow\downarrow}^R(E_0)} \quad (11)$$

$$C_o(E_0) = \frac{2VA(E_0) - g_{\uparrow}^R(E_0) - g_{\downarrow}^R(-E_0)}{\pi f_{\uparrow\downarrow}^R(E_0)} \quad (12)$$

and $A(E_0) = f_{\uparrow\downarrow}^R(E_0)^2 + g_{\uparrow}^R(E_0)g_{\downarrow}^R(-E_0)$. In these expressions, the bare substrate Green function f and g are defined according to (6). Thus the even/odd- ω components of the LDOS defined as $\rho_{\text{even/odd}}(\omega) = (\rho(\omega) \pm \rho(-\omega))/2$ are directly proportional to the imaginary part of odd/even-frequency pairings respectively:

$$\rho_{\text{even/odd}}(\omega) = C_{e/o}(E_0) \times \Im F_{\text{odd/even}}^R(\omega). \quad (13)$$

We have therefore derived a general proportionality relation between the even- ω part of the LDOS and the imaginary part of the odd- ω anomalous pairing function. This relationship has the strong physical consequence that as soon as there exists some in-gap YSR state, there is a local odd- ω pairing around the impurity site. Note that this is a direct consequence of the magnetic impurity locally breaking time-reversal symmetry.

The proportionality coefficient does in general depend on the way the substrate is modeled. However, in most physically relevant cases the Fermi Energy of the substrate is the largest energy scale and the normal DOS can be approximated by its value at the Fermi Energy, ν_0 . Considering that we have a single YSR in-gap bound state, its energy E_0 is given by

$$E_0 = \Delta \frac{1 - \alpha^2 + \beta^2}{\sqrt{(1 - \alpha^2 + \beta^2)^2 + 4\alpha^2}}, \quad (14)$$

where $\alpha = \pi\nu_0 J$ and $\beta = \pi\nu_0 V$ [21]. This allows one to easily express the proportionality coefficient,

$$\begin{aligned} C_e(E_0) &= -\frac{2}{\Delta} [E_0 + \pi J \nu_0 \sqrt{\Delta^2 - E_0^2}] \\ &= -\frac{2}{\pi} \frac{1 + \beta^2 + \alpha^2}{\sqrt{(1 - \alpha^2 + \beta^2)^2 + 4\alpha^2}}. \end{aligned} \quad (15)$$

It is important to emphasize here that, on the contrary to $C_o(E_0)$ which vanishes in the case of a pure magnetic impurity ($V = 0$), $C_e(E_0)$ never vanishes. We can thus always evaluate $\Im F_{\text{odd}}^R(\omega) = \rho_{\text{even}}(\omega)/C_e(E_0)$. Notice that $\pi|C_e^{-1}|/2 \in [0, 1]$. Given $\rho_{\text{even}}(\omega)$, the odd- ω pairing function is maximal for $|C_e^{-1}| = 2/\pi$ which is reached for $\beta = 0$. This corresponds to a pure magnetic impurity. The opposite limit $|C_e^{-1}| \rightarrow 0$ is reached only for $\beta^2 = \alpha^2 + 1 = \infty$ which are unphysical values. Even for extremely large values of $\alpha, \beta \sim 4$, we can still obtain a lower bound for $|\Im F_{\text{odd}}^R(\omega)|$. Inside the gap, the pairing thus has the same order of magnitude as the LDOS.

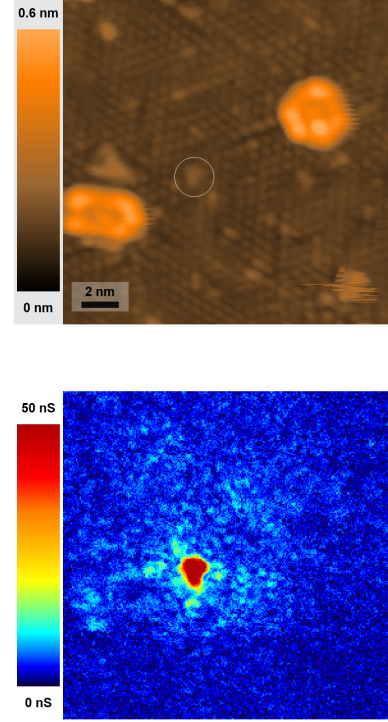


FIG. 1: Upper panel: scanning tunnelling microscopy image of the Pb monolayer where a magnetic defect is present as a triangular protrusion. Lower panel: corresponding conductance map measured at the Fermi level by scanning tunnelling spectroscopy at 320mK.

Protocol to extract odd- ω pairing. Let us now provide an efficient protocol to extract the imaginary part of the local odd frequency pairing function $\Im F_{\text{odd}}^R(\omega)$ around the impurity from LDOS spectroscopic measurements performed in the tunneling regime. The differential conductance spectrum dI/dV measured locally corresponds to the convolution of the local density of state $\rho(\omega)$ with the derivative of the Fermi-Dirac distribution at the experimental temperature. Once $\rho(\omega)$ is measured, we normalize it in units of the normal-state DOS at the Fermi-level denoted ν_0 . In order to extract a reliable estimate of $\Im F_{\text{odd}}^R(\omega)$ we perform some simple data analysis. To do so, we follow [51] and assume that the YSR is well approximated by the following retarded Green function

$$\hat{G}(\omega) = \frac{1}{\omega + i\eta - E_0} \cdot \begin{bmatrix} u^2 & uv \\ uv & v^2 \end{bmatrix}, \quad (16)$$

$$u^2, v^2 = 2\pi\nu_0\Delta \frac{1 + (\alpha^2 \pm \beta^2)}{((1 - \alpha^2 + \beta^2)^2 + 4\alpha^2)^{3/2}}, \quad (17)$$

where, u and v are the electron and hole components of the YSR state. This expression is obtained from the exact solution of the Dyson equation after a first-order expansion around energy E_0 [51]. The phenomenological parameter η introduced here takes into account the

broadening of the YSR peaks due to relaxation. It can be related to Γ up to some renormalization coefficient. With this expression of the Green function, one then obtains

$$\rho(\omega) = \frac{\eta u^2/\pi}{(\omega - E_0)^2 + \eta^2} + \frac{\eta v^2/\pi}{(\omega + E_0)^2 + \eta^2}. \quad (18)$$

Using this expression to fit the experimental data, we can extract the parameters u^2 , v^2 and the inverse lifetime η . With these values at hand, one obtains the coefficient $C_e(E_0) = -\frac{u^2+v^2}{\pi uv}$ and thus $\Im F_{odd}^R(\omega) = \rho_{even}(\omega)/C_e(E_0)$. Note that $\Re F_{odd}^R(\omega)$ can be obtained by the Kramers-Kronig relation.

Application to magnetic impurities in a Pb/Si(111) substrate. We apply the previous protocol to extract $\Im F_{odd}^R(\omega)$ based on local density of states (LDOS) measurements, and test it on STS data of magnetic impurities immersed in a Pb/Si(111) monolayer. The Pb monolayer corresponds to a nominal coverage of 4/3 with the stripe incommensurate reconstruction. This Pb monolayer was shown to be superconducting below 1.8K [52]. This system does not show any in-gap states in the presence of a strong non-magnetic disorder as expected for a s-wave superconductor [53]. However, in presence of magnetic defects YSR states appear and manifest by huge pairs of conductance peaks in a well-defined gap [54]. Moreover, we always observe a single pair of conductance peak meaning that only one YSR state is present or at least that the eventual multiplet is degenerated up to the experimental resolution. Note that as the Pb monolayer is a 2D superconductor, the YSR states extend very far from the impurities (typically tens of nanometers) [55]. However, we focus here on spectra taken on top of the impurities. The upper panel of Fig. 1 shows a scanning tunnelling microscopy image of the Pb monolayer where a magnetic defect is present as a triangular protrusion, the corresponding conductance map measured at the Fermi level by STS at 320mK is shown in the lower panel of Fig. 1. One can see a red spot on top of the defect that corresponds to a very strong YSR state, that is surrounded by a speckle like pattern due to the scattering of the decaying YSR wave function scattered by the atomic disorder of the monolayer.

The upper panel of Fig. 2 shows a spectrum (black curve) taken far from the impurity that corresponds to a BCS gap of 0.38 meV with a broadening $\Gamma_{Dynes} \propto \Gamma = 0.004$ meV convoluted by a thermal broadening due to the finite temperature of 320 mK. By contrast a spectrum taken on top of the impurity (blue dots) exhibits a strong pair of YSR peaks in the gap (see Fig. 2). From the differential conductance, we can then extract the LDOS $\rho(\omega)$ at the impurity site which is represented in the lower panel of Fig. 2. We checked that the reconvoluted DOS matches perfectly the original spectrum. We then fit the data with the equation defined in (18). The fitted results for $\rho(\omega)$ are displayed in the middle panel of Fig. 2 and show a good agreement with the data. In order not to

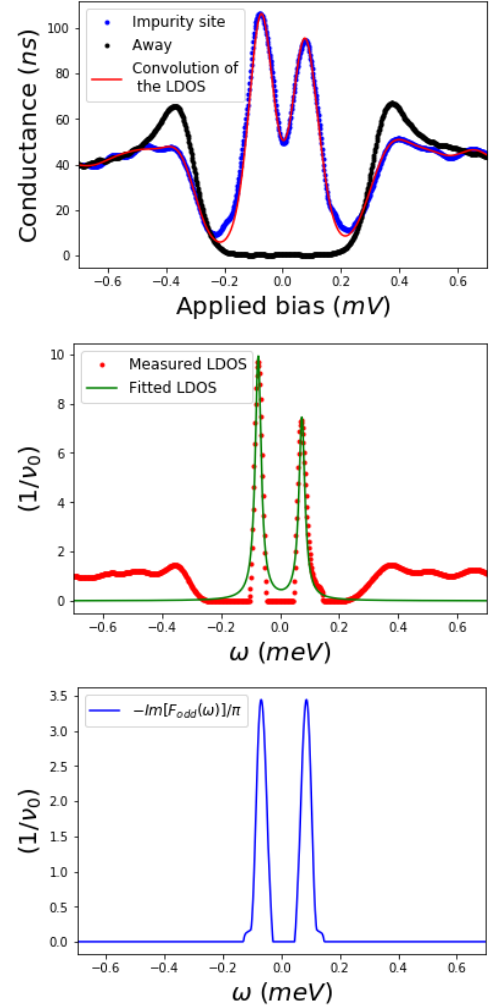


FIG. 2: Upper panel: Measured differential conductance at the impurity site (blue dots), far away from the impurity (black dots) and the one obtained from convolution of the extracted LDOS with the Fermi-Dirac derivative (red line). Middle panel: LDOS obtained after deconvolution taking into account the finite temperature $T=320$ mK (red dots). Fitted LDOS with the set of parameters: $E_0 = 0.074$ meV, $\eta = 0.012$ meV, $u^2 = 0.28\nu_0$, $v^2 = 0.38\nu_0$ (green curve). Bottom panel: Extracted $-\Im F_{odd}^R(\omega)/\pi$ at the position of the impurity (the value are relevant only inside the gap).

overestimate $\Im F_{odd}^R(\omega)$, we always take into account the parameters set which maximizes the ratio v^2/u^2 . With the parameters u, v, η at hand, we have thus access to $C_e(E_0)$. We have plotted the so-obtained $-\Im F_{odd}^R(\omega)/\pi$ in the lower panel of Fig. 2. Notice that its amplitude is comparable to $\rho(\omega)$. We have applied this procedure to other sets of YSR states, fully confirming the results of this analysis (see supplementary material [56]).

Conclusion. In conclusions, we show that an isolated magnetic impurity in an s-wave superconductor generates local pairing correlations which are odd in frequency. We provided a protocol to extract these anomalous pairing

functions from STS measurements and apply it to data taken from a Pb/Si(111) monolayer with magnetic impurities. Our theoretical/experimental analysis finally proves the occurrence of odd-frequency pairing in the simplest magnetic-superconductor hybrid system.

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Note added: As we were finalizing the final writing of this manuscript, we learned about the theoretical work of D. Kuzmanovski *et al.* [57] which has partial overlap with our work and reaches similar conclusions.

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