Enhanced Antiferromagnetic Exchange between Magnetic Impurities in a Superconducting Host

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It is generally believed that superconductivity only weakly affects the indirect exchange between magnetic impurities. If the distance r between impurities is smaller than the superconducting coherence length ($r \lesssim \xi$), this exchange is thought to be dominated by Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions, identical to the those in a normal metallic host. This perception is based on a perturbative treatment of the exchange interaction. Here, we provide a nonperturbative analysis and demonstrate that the presence of Yu-Shiba-Rusinov bound states induces a strong $1/r^2$ antiferromagnetic interaction that can dominate over conventional RKKY even at distances significantly smaller than the coherence length ($r \ll \xi$). Experimental signatures, implications, and applications are discussed.

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Understanding the interactions between magnetic impurities (localized spins) in a metallic host represents an important question at the interface of fundamental and applied science [1–5]. While spins always interact with one another via their intrinsic dipolar interaction, in a metal, their mutual interaction with conduction electrons can significantly enhance the effective interactions. For simple metals, this results in the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction [2–4]—a coupling mechanism between magnetic moments in which one impurity partially polarizes the spin of conduction electrons; the second impurity then interacts with the spin density of the itinerant electrons, thereby inducing an effective long-range interaction. One of the crucial predictions of RKKY is the oscillatory sign of the exchange interaction, a feature which underlies giant magnetoresistance [6,7].

More recently, significant effort has been devoted to understanding magnetic impurities on the surface of superconducting metals [5,8–16]. This owes, in part, to experimental advances in single adatom control, which have enabled the observation of locally modified electronic properties and raise the tantalizing prospect of atom-by-atom construction of magnetic nanostructures [17–19]. Moreover, interactions between such impurities may play a role in explaining low-frequency flux noise in Josephson circuits [20,21].

The effect of superconductivity on RKKY interactions is well established at lowest-order perturbation theory (Born approximation) in the exchange interaction between the localized and itinerant spins. In particular, the suppressed spin susceptibility in the superconducting ground state modifies the interimpurity interaction to become purely antiferromagnetic when the separation between the impurities exceeds the superconducting coherence length ($r \gtrsim \xi$); at such distances however, the strength of this antiferromagnetic exchange is exponentially small in the separation r. On the other hand, for impurities separated by distances $r < \xi$, conventional RKKY dominates the effective interaction and superconductivity yields only a weak antiferromagnetic correction [22–24]. Crucially, this perturbative treatment neglects the formation of so-called Yu-Shiba-Rusinov (YSR) bound states—localized electronic states that arise near a magnetic impurity.

In this Letter, we show that by tuning the energy of YSR states close to the middle of the superconducting gap, one may substantially enhance the antiferromagnetic contribution stemming from the indirect spin exchange, allowing it to dominate over conventional RKKY even at distances $r \lesssim \xi$ [25–27]. When two magnetic impurities are brought near one another, their associated YSR states hybridize in a spin-dependent fashion, yielding an effective interaction. That one might expect such an interaction to dominate over RKKY results, in part, from the strong localization of the YSR state around the impurity, directly contrasting with the delocalized scattering states that mediate RKKY. This localization implies that quasiparticles bound to the YSR states are more strongly coupled to the impurity and therefore might be expected to mediate stronger exchange.

The key ideas underlying our derivation are illustrated in Fig. 1. We begin by considering a BCS superconductor with Hamiltonian,

$$H_0 = \sum_{\mathbf{k},\sigma} \epsilon_{\mathbf{k}} c^\dagger_{\mathbf{k},\sigma} c_{\mathbf{k},\sigma} + \Delta \sum_{\mathbf{k}} [c^\dagger_{\mathbf{k}\uparrow} c^\dagger_{-\mathbf{k}\downarrow} + c_{-\mathbf{k}\downarrow} c_{\mathbf{k}\uparrow}]. \quad (1)$$

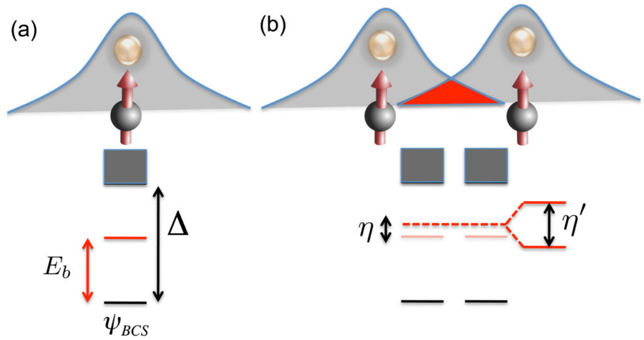

FIG. 1 (color online). (a) Schematic illustration of a magnetic impurity which binds a localized electronic YSR state. The associated spectrum is shown below, with the BCS ground state $\psi_{\rm BCS}$ separated from excited states by Δ. There exists a single midgap YSR state of energy E_b. (b) When two impurities are separated by distances $r < \xi$, their YSR states overlap and hybridize. This hybridization causes both an overall energy shift η and a splitting η'.

The associated spectrum (Fig. 1) depicts the BCS ground state, $\psi_{\rm BCS}$, separated from excited states by the superconducting gap Δ. In the presence of a spin impurity whose contact exchange interaction is of strength J, an excited-state electron can lower its energy below the superconducting gap by aligning its spin opposite the direction of the impurity. Treating the spin impurity classically yields the existence of a localized bound state (YSR state) of energy [25–27],

$$E_b = \Delta \frac{1-(\pi S N_0 J/2)^2}{1+(\pi J S N_0/2)^2} = \Delta \frac{1-\beta^2}{1+\beta^2}, \tag{2}$$

where N_0 is the normal state DOS at the Fermi energy. For pure exchange scattering, the YSR energy is conveniently reexpressed in terms of a phase shift $\tan(\delta) \equiv \beta = \pi S N_0 J/2$, wherein $E_b = \Delta\cos(2\delta)$. This latter relation between E_b and δ is valid beyond the classical magnetic impurity approximation, which is used in relating β to J [28–31]. In the absence of superconductivity, quantum spin fluctuations result in the Kondo effect which renormalizes the exchange interaction between the impurity and itinerant electrons [32] at energies D, low compared to the Fermi energy E_f. Within perturbation theory, the renormalized exchange is given by $N_0 J(D) = N_0 J[1 + N_0 J \ln(E_f/D)]$; the comparison between the two terms here defines the so-called Kondo temperature, $T_K \propto \exp(1/N_0 J)$. A more detailed renormalization group (RG) treatment allows one to extend this perturbative result to the scaling regime [33], wherein $N_0 J(D) \approx 1/\ln(D/T_K)$ at $D \gtrsim T_K$. For a magnetic impurity in a superconductor with $\Delta \gg T_K$, the RG stops at $D \sim \Delta$. This yields the replacement of $N_0 J \to 1/\ln(\Delta/T_K)$ in β, under the assumption that $\beta \lesssim 1$ [32].

The characteristic wave function of the YSR state is localized around the magnetic impurity and takes the form, $\phi(\mathbf{r}) \sim (1/\mathbf{r}) e^{-\mathbf{r}/\xi|\sin(2\delta)|}$ [5]. For two impurities separated by distances $r \gg \xi$, the overlap between their associated YSR states is exponentially suppressed. However, for distances $r < \xi$, the YSR states of the two impurities hybridize, causing both an overall energy shift η and a splitting η', as depicted in Fig. 1(b). Crucially, the overall energy shift η depends on whether the impurity spins are aligned or antialigned; in particular, only in the antialigned case is it possible for a pair of YSR states to become virtually occupied by a Cooper pair from the superconducting condensate. This provides a natural intuition for our result: the effective spin-spin interaction manifests as a consequence of the spin dependence in η.

With this intuition in mind, we now begin by considering the total energy associated with a pair of magnetic impurities (located at r_L and r_R) in a superconductor. We treat the impurities as classical spins parallel to the $\hat{z}$ axis (which defines the direction in which the impurities are either aligned or antialigned). The interaction Hamiltonian between the localized impurity and the itinerant electrons is then given by

$$H_{\rm int} = J \sum_\sigma \int d\mathbf{r}\sigma[S_L f(\mathbf{r}-\mathbf{r}_L) c^\dagger_\sigma(\mathbf{r}) c_\sigma(\mathbf{r}) + S_R f(\mathbf{r}-\mathbf{r}_R) c^\dagger_\sigma(\mathbf{r}) c_\sigma(\mathbf{r})], \tag{3}$$

where $S_{L(R)}$ is the spin of the left (right) impurity and $f(\mathbf{r})$ characterizes the spatial form of the impurity potential [34,35]. In momentum space, $H_{\rm int} = J\sum_\sigma \int d\mathbf{k} d\mathbf{k}'\sigma\times [S_L e^{i(\mathbf{k}-\mathbf{k}')r_L}\tilde{f}_{\mathbf{k},\mathbf{k}'} + S_R e^{i(\mathbf{k}-\mathbf{k}')r_R}\tilde{f}_{\mathbf{k},\mathbf{k}'}] c^\dagger_{\sigma,\mathbf{k}} c_{\sigma,\mathbf{k}'}$, where $\tilde{f}$ is the Fourier transform of the potential. As is conventional [36], we now define a Nambu spinor, $\Psi_\mathbf{k} = (c_{\uparrow,\mathbf{k}}, c^\dagger_{\downarrow,-\mathbf{k}})$, wherein, $H_0 = \int d\mathbf{k} \Psi^\dagger_\mathbf{k}[\epsilon_\mathbf{k}\tau^z + \Delta\tau^x]\Psi_\mathbf{k}$ (τ are Pauli matrices acting in particle-hole space). Similarly, the interaction becomes,

$$H_{\rm int} = J \int d\mathbf{k} d\mathbf{k}' \Psi^\dagger_\mathbf{k}[S_L e^{i(\mathbf{k}-\mathbf{k}')r_L}\tilde{f}_{\mathbf{k},\mathbf{k}'} + S_R e^{i(\mathbf{k}-\mathbf{k}')r_R}\tilde{f}_{\mathbf{k},\mathbf{k}'}]\Psi_{\mathbf{k}'} + E_0, \tag{4}$$

where $E_0 = -J\int d\mathbf{k}\tilde{f}_{\mathbf{k},\mathbf{k}}[S_L + S_R]$ arises from anticommutation.

Combining the bare BCS Hamiltonian and the interactions yields, $H_T = H_0 + H_{\rm int}$, which we diagonalize utilizing a Bogoliubov transformation, $d^\dagger_n = \int d\mathbf{k}(u_{n,\mathbf{k}}\psi^\dagger_{\uparrow,\mathbf{k}} + v_{n,\mathbf{k}}\psi^\dagger_{\downarrow,\mathbf{k}})$, yielding

$$H_T = \sum_n \varepsilon_n d^\dagger_n d_n - \frac{1}{2}\sum_n \varepsilon_n = \sum_n \varepsilon_n\left(d^\dagger_n d_n - \frac{1}{2}\right). \tag{5}$$

The total energy of the ground state is thus given by

$$E_{\rm tot} = -\frac{1}{2}\sum_n |\varepsilon_n| = E_V - \frac{1}{2}\int d\epsilon|\epsilon|\delta\rho(\epsilon), \tag{6}$$

where E_V characterizes the energy of the system in the absence of an impurity. Here, $\delta\rho(\epsilon)$ represents the change in the total density of states as a result of the impurities and includes contributions from both continuum electronic states above the gap as well as the discrete YSR states. The effective exchange interaction, $I(\mathbf{r})$, between two impurities can be expressed in terms of changes to the DOS depending on whether the impurities are aligned or antialigned,

$$I(\mathbf{r}) = E_{\rm tot}^{\uparrow,\downarrow} - E_{\rm tot}^{\uparrow,\uparrow} = -\frac{1}{2}\int d\epsilon|\epsilon|[\delta\rho_{\uparrow,\downarrow}(\epsilon) - \delta\rho_{\uparrow,\uparrow}(\epsilon)]. \quad (7)$$

To calculate changes in the DOS, we compute $\delta\rho(\epsilon) = -(1/\pi)\mathrm{Im}\{\mathrm{Tr}[G_{\mathbf{k},\mathbf{k}'}(z) - G_{\mathbf{k}}^{(0)}(z)]\}$, where $z = \epsilon + i0^+$, $G_{\mathbf{k}}^{(0)}(z) = [z - (\epsilon_{\mathbf{k}}\tau^z + \Delta\tau^x)]^{-1}$ is the bare BCS Green's function, and $G_{\mathbf{k},\mathbf{k}'}(z)$ is the perturbed Green's function. Since translational invariance is broken by the magnetic impurities, the perturbed Green's function depends on two momenta, $\mathbf{k}$ and $\mathbf{k}'$. Working within the T-matrix formalism [5],

$$G_{\mathbf{k},\mathbf{k}'}(z) = G_{\mathbf{k}}^{(0)}(z) + G_{\mathbf{k}}^{(0)}(z)T_{\mathbf{k},\mathbf{k}'}G_{\mathbf{k}'}^{(0)}(z), \quad (8)$$

where $T_{\mathbf{k},\mathbf{k}'}$ is the T matrix. Applying a Dyson expansion to the T matrix (see the Supplemental Material [37]), one finds that

$$\begin{aligned}\delta\rho(\epsilon) &= -\frac{1}{\pi}\mathrm{Im}\{\mathrm{Tr}[G_{\mathbf{k}}^{(0)}(z)T_{\mathbf{k},\mathbf{k}'}G_{\mathbf{k}'}^{(0)}(z)]\} \\ &= -\frac{1}{\pi}\mathrm{Im}\{\mathrm{Tr}[JS\Pi(1 - JSG)^{-1}]\}, \quad (9)\end{aligned}$$

where Π, G, and S are 4×4 matrices (in the tensor product space of particle-hole and left-right position) given by

$$\Pi_{ll'}(z) = \int d\mathbf{k}G_{\mathbf{k}}^{(0)}(z)G_{\mathbf{k}}^{(0)}(z)e^{i\mathbf{k}(\mathbf{r}_l - \mathbf{r}_{l'})}, \quad (10)$$

$$G_{ll'}(z) = \int d\mathbf{k}G_{\mathbf{k}}^{(0)}(z)e^{i\mathbf{k}(\mathbf{r}_l - \mathbf{r}_{l'})}, \quad (11)$$

$$S_{ll'} = S_l\delta_{ll'} \otimes \tau^0. \quad (12)$$

Here, τ^0 represents the identity matrix in particle-hole space and l, l' run over $\{L, R\}$, indexing the left or right impurity; we emphasize that the above formalism can naturally be extended to multiple ($N > 2$) impurity calculations [34,35].

We begin by considering the case of weakly bound YSR states ($J \ll 1$) and expand Eq. (9) to second order in the exchange coupling, $\mathrm{Tr}[JS\Pi(1 - JSG)^{-1}] \approx \mathrm{Tr}[J^2S\Pi SG]$. Evaluating this perturbative expression results in the following superconducting RKKY exchange between the magnetic impurities:

$$\begin{aligned}I(\mathbf{r}) = &\frac{E_f\beta^2}{\pi(k_f r)^3}\cos(2k_f r)e^{-(2r/\xi)}F_1\left[\frac{2r}{\xi}\right] \\ &+ \frac{\Delta\beta^2}{(k_f r)^2}\sin^2(k_f r)e^{-(2r/\xi)}F_2\left[\frac{2r}{\xi}\right]. \quad (13)\end{aligned}$$

Here, k_f is the Fermi momentum, $r = |\mathbf{r}_L - \mathbf{r}_R|$ is the distance between the spins and $F_1[\alpha] = \alpha\int_0^\infty dx e^{-\alpha(\sqrt{x^2+1}-1)}$, $F_2[\alpha] = (2/\pi)\int_0^\infty dx(e^{-\alpha(\sqrt{x^2+1}-1)}/(x^2+1))$ are dimensionless integrals. The first term represents the bare superconducting RKKY interaction, while the second represents an additional antiferromagnetic correction. Although this second term scales as $1/r^2$, it is weaker by a factor of Δ/E_f and only dominates over bare superconducting RKKY at distances $r \gg E_f/(\Delta k_f) \sim \xi$, by which time the entire exchange integral $I(\mathbf{r})$ is exponentially suppressed. The above perturbative result is consistent with previous calculations which utilize the Kubo formula to compute the exchange interaction from the magnetization response [22–24,37].

Returning to the interpretation of the exchange energy in terms of changes to the density of states [Eqs. (7) and (9)], we recall that the effective exchange contains two contributions, one from continuum electronic states and the other from discrete YSR states. One might expect that, being only weakly bound, the YSR states should induce a contribution which decays more slowly than $e^{-(2r/\xi)}$. However, we find that at $\mathcal{O}(J^2)$, the tail of the YSR contribution exactly cancels with a portion of the continuum contribution to yield the perturbative expression found in Eq. (13).

Moving beyond the perturbative limit, as J increases, the energy of the YSR bound state decreases (approaching the middle of the superconducting gap) and the relative strength of the continuum and YSR contributions change. In particular, one might expect the YSR contribution to dominate for deeply bound states for two reasons: first, modifications to the bulk DOS will become weaker (since the bound state is further from the bottom of the band), and second, YSR hybridization with the superconducting condensate will become stronger as $E_b \to 0$. This second point suggests that the energy shift η has the potential to develop a singular contribution, arising from the $|\epsilon|$ in Eq. (6) near $\epsilon \approx 0$; thus, any singular contribution to the exchange interaction can only arise from the low energy YSR states.

To see these effects explicitly, we now compute the bound state energies as a function of impurity separation. This corresponds to a direct calculation of the discrete YSR contribution to Eq. (7). The YSR bound state energies can be computed from poles of $\mathrm{Tr}[G_{\mathbf{k},\mathbf{k}'}(z)]$. More explicitly, E_b is determined by

$$F(E_b) \equiv \mathrm{Det}[1 - SG(E_b)] = 0. \quad (14)$$

In the limit, $k_f r \gg 1$, one can consider the hybridization of the isolated YSR bound states to obtain perturbative corrections to the YSR energies. We derive an analytic approximation for solutions of Eq. (14) in the case of both parallel and antiparallel impurities (see the Supplemental Material [37]). By subtracting the bare YSR energy

[Eq. (2)], this allows us to compute the spin-dependent total energy shift η. Our perturbative expansion is in the parameter η/E_b and remains valid so long as the energy shift is small relative to the bare YSR energy [see Eq. (16) and below for a discussion of validity].

We first consider the case of antiparallel impurities where symmetry allows us to directly expand around the bare YSR energy, $F(E_b) + \eta_{\uparrow\downarrow} F'(E_b) = 0$. A straightforward but tedious calculation then yields the leading term in $1-\beta$ as $\eta_{\uparrow\downarrow} = \Delta(1/1-\beta)(\cos^2(k_f r)/2(k_f r)^2)e^{-(2r/\xi)}$. In the case of parallel spins, the situation is slightly more complicated since one must extract the total shift by averaging the split energies [Fig. 1(b)]. This requires expanding to third order, $F(E_b) + \eta_{\uparrow\uparrow} F'(E_b) + \frac{1}{2}\eta_{\uparrow\uparrow}^2 F''(E_b) + \frac{1}{6}\eta_{\uparrow\uparrow}^3 F'''(E_b) = 0$ and results in a non-singular shift, $\eta_{\uparrow\uparrow} = -(\Delta/2)(\cos(k_f r)/(k_f r)^2)e^{-(2r/\xi)}$, as $\beta \to 1$(see the Supplemental Material [37]).

The YSR contribution to the exchange, $I(\mathbf{r})$, is given by $J_{\mathrm{YSR}} = \eta_{\uparrow\downarrow} - \eta_{\uparrow\uparrow}$ [37]. Crucially, as the bound state energy approaches the middle of the superconducting gap ($E_b \to 0$, $\beta \to 1$), J_{YSR} is dominated by the singular contribution in $\eta_{\uparrow\downarrow}$ yielding

$$J_{\mathrm{YSR}} = \Delta \frac{1}{1-\beta} \frac{\cos^2(k_f r)}{2(k_f r)^2} e^{-(2r/\xi)}, \tag{15}$$

which exhibits a resonant enhancement of the form $1/1-\beta$. This resonant enhancement has an intuitive explanation. It arises from the hybridization of a pair of YSR states with the superconducting condensate; more specifically, when the impurities are antialigned, this hybridization occurs as a result of the conversion of a Cooper pair from the condensate into a pair of electrons in the YSR states. Conceptually, this intuition is somewhat related to the superexchange between magnetic ions; indeed, owing to Pauli-blocking, such superexchange interactions are also typically antiferromagnetic in nature [39,40]. At a heuristic level, coupling to the condensate takes the form $\Delta U(r) c^\dagger_{L,\uparrow} c^\dagger_{R,\downarrow}$, where $U(r) = \cos(k_f r)/(k_f r)$ characterizes the overlap between the bound states. While the ground state energy correction stemming from this coupling is generally suppressed by an energy denominator $2E_b$, as β approaches unity, E_b approaches zero, leading to the observed resonant enhancement.

The physical limit of the enhancement of this purely antiferromagnetic contribution is set by the condition that the YSR energies have not crossed zero, which in effect, would signify a parity changing transition. This condition also represents the regime of validity for J_{YSR} as derived from the expansion of Eq. (14). In combination with the constraint that J_{YSR} dominates over bare RKKY interactions, we obtain a double-sided inequality,

$$k_f r > \frac{1}{1-\beta} > \frac{\xi}{r}. \tag{16}$$

By stark contrast to the perturbative limit, where the superconducting correction dominates only at distances $r \gg \xi$, here, we find that the antiferromagnetic J_{YSR} exchange can prevail at $r \sim \sqrt{\lambda_f \xi} \ll \xi$ and reaches a maximum ($\sim\Delta/k_f\xi$) at such distances.

Discussion.—Inspection reveals that the YSR-induced interaction strength, $J_{\mathrm{YSR}} = \eta_{\uparrow\downarrow} - \eta_{\uparrow\uparrow}$ scales as $\sim(1/r^2)$, exhibiting a weaker decay than conventional metallic RKKY interactions. We note that this power law is in agreement with the perturbative superconducting correction in Eq. (13); as expected, for small β, our full nonpertubative calculation matches the perturbative results (see the Supplemental Material [37]). In comparison to bare RKKY interactions, one important qualitative observation is that, while oscillatory in nature, J_{YSR} does not vary between ferromagnetic and antiferromagnetic couplings. The antiferromagnetic nature of the superconducting YSR correction results from the fact that coupling to the condensate can only occur for antialigned impurities.

For small impurity separation and weakly bound YSR states, the magnitude of the RKKY interaction dominates over J_{YSR}. However, as illustrated in Fig. 2, for bound state energies close to the middle of the gap, resonant enhancement enables $J_{\mathrm{YSR}} > J_{\mathrm{RKKY}}$ at distances well below the coherence length; the dominance of this antiferromagnetic exchange is further highlighted by the weaker power-law decay as a function of r. This effect will be especially pronounced for superconductors with relatively large coherence lengths.

To observe the resonant enhancement of J_{YSR} requires a system where the coupling strength between the impurity spin and the superconductor can be tuned continuously. In principle, any low-density system with a tunable DOS can provide a natural mechanism for controlling the exchange constant via a gate voltage. An example of such a scenario is found in graphene [41], where the exchange coupling of magnetic defects can be altered by simply changing the carrier density. In combination with demonstrations of proximity-induced superconductivity [42], this suggests that graphene in contact with a superconductor may represent a promising system with which to realize tunable-energy YSR states. Such a system naturally possesses a

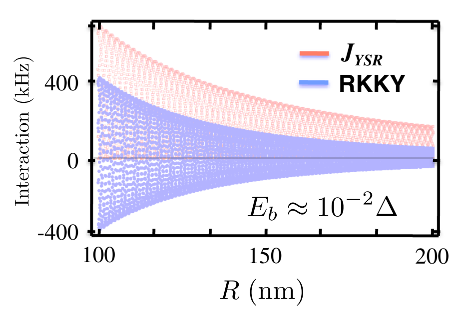

FIG. 2 (color online). For concreteness, all plots are calculated using actual parameters for superconducting aluminum, with $E_f = 11.7$ eV, $k_f = 20.1\ \mathrm{nm}^{-1}$, $N_0 = 35\ \mathrm{eV/nm}^3$, and $\xi = 1.6\ \mu$m [38]. Comparison between bare RKKY and the J_{YSR} for $E_b \sim 10^{-2}\Delta$. Resonant enhancement enables J_{YSR} to dominate at distances $r \ll \xi$.

large coherence length since the Fermi velocity remains substantial even at low carrier densities. Interestingly, it may also be possible to further enhance the effects of an applied gate voltage by separating the graphene from the superconductor via a layer of semiconductor such as MoS_2 [43,44].

In summary, working beyond the Born approximation, we have derived an enhanced antiferromagnetic exchange between magnetic impurities embedded in a superconducting host. Such an interaction provides a fundamental limit to the formation of spin-helical order [45,46], which underlies recent proposals for the observation of topological superconductivity and Majorana bound states in a 1D YSR impurity chain [47–49]. Although our results are formulated within the treatment of classical spins, such a description is consistent for high-spin magnetic ions such as those currently used in experiments (e.g., Gd, Mn, Cr) [17,18]. Finally, our Letter is complementary and in parallel with recent numerical renormalization group studies on the two-impurity YSR phase diagram [50].

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