Giant superconducting proximity effect on spintronic anisotropy

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We investigate theoretically the interplay of proximity effects due to the presence of a superconductor and normal ferromagnetic leads on the formation of the spintronic quadrupolar exchange field in a large-spin molecule. We show that the spintronic anisotropy can be enhanced by a few orders of magnitude, and tuned by changing the strength of coupling to the superconductor. Especially large anisotropy is generated in the vicinity of the charge parity changing transition of the molecule. We also provide predictions of measurable spectral properties being the hallmarks of these phenomena.

Introduction.—Devices based on large-spin ($S \geq 1$) nanoscopic systems (e.g., adatoms [1, 2]; molecules [3–5] or engineered quantum-dot structures [6, 7]) hold promise to become crucial components of electronic and spintronic circuits. The behavior of a spin-degree ground multiplet in such a device is captured by the effective Hamiltonian \[ H_{\text{eff}} = BS_z + DQ_{zz}. \] Here, the first term represents the effect of a magnetic field $B$ on the component of the total spin along the $z$-axis, $S_z$. The second term, on the other hand, describes uniaxial magnetic anisotropy (quantified by a constant $D$) associated with a diagonal, $z$th component of the spin-quadrupole moment tensor, $\tilde{Q}_{zz} \equiv \tilde{S}_z^2 - S(S+1)/3$ [9]. Importantly, for $D < 0$, this anisotropy leads to formation of the energy barrier $\Delta \mathcal{E}$ for spin reversal between two metastable states $\{\pm S_z\}$, see Fig. 1(a)—the prerequisite for a magnetic bistability, indispensable for applications in information-processing technologies [10–12]. In an electronic circuit, $B$ corresponds then to an externally applied magnetic field, while $D$ stems from the intrinsic spin-orbit interaction in the device [13]. However, in a spintronic circuit, where electronic transport is spin-polarized, both $B$ and $D$ can be entirely generated spintronically in the form of effective dipolar [14–17] and quadrupolar [18–20] exchange fields, respectively—here, also referred to as ‘spintronic fields’. As a result, both the magnetic anisotropy of the device and its spin state can be on demand controlled in a fully electrical manner. The success of this approach hinges on the possibility of tuning and inducing significantly large spin-reversal barriers, which in the case of intrinsic magnetic anisotropy can be relatively small and hardly fine-controllable [21].

In this Rapid Communication, we propose how to boost this barrier by orders of magnitude and control it with the aid of the superconductor (SC) proximity effect. Hybrid nanostructures involving molecules or quantum dots attached to normal and SC electrodes are nowadays attracting a considerable attention [22, 23]. Such systems enable the exploration of Yu-Shiba-Rusinov states [24–26] by means of Andreev spectroscopy [27–29] and have proved useful for solid-state-based generation of entangled states by splitting the Cooper pairs [30, 31]. Moreover, the superconducting proximity can strongly influence the transport properties of hybrid devices, especially in the strong correlation regime, and generate a quantum critical behavior [32–34]. Here, we focus on a new aspect of SC proximity, namely, we unveil its beneficial impact on spintronic magnetic anisotropy in molecular systems.

To demonstrate this impact, we consider a prototypical hybrid device consisting of a large-spin, intrinsically isotropic molecule [35] embedded into a magnetic tunnel junction and proximized by a SC, see Fig. 1(b). Employing the numerical renormalization group (NRG) approach [36], electronic transport through such a device is investigated reliably in a broad range of tunnel couplings between the molecule and the junction. We show that the spintronic anisotropy can be controllably tuned by changing the strength of coupling to the superconductor. In particular, we predict an enhancement of anisotropy by a few orders of magnitude due to the SC proximity for parameters tuned around the transition between two different ground states of the molecule. Our work opens, thus, new perspectives for manipulating the properties of magnetic molecules, which are indispensable for applications in future information storage technologies and further applications in future information storage technologies and spintronics.

Figure 1. Example of an energy spectrum for a model system with the ground spin-multiplet $S = 3/2$ and uniaxial spintronic anisotropy $D < 0$. (b) Schematic of a SC-proximized large-spin molecule with a molecular level (ML) occupied by a single electron connected to two metallic ferromagnetic leads. For details see the main text.
molecular spintronics.

**Theoretical description and method.**—A large-spin molecule is modeled as composed of two parts [37]: a magnetic core (represented as an effective spin $\hat{S}_c$) and a single molecular level (ML), which mediates transport of electrons between the leads. The ML can be occupied by up to two electrons, which is accounted for by the Hamiltonian $\hat{H}_{\text{ML}} = \varepsilon \sum_{\sigma} \hat{n}_\sigma + U \hat{n}_\uparrow \hat{n}_\downarrow$. Here, $\hat{n}_\sigma = \hat{d}^\dagger_\sigma \hat{d}_\sigma$ is the occupation operator for an electron of spin $\sigma$, whereas $\varepsilon$ and $U$ stand for single-occupation and Coulomb energies, respectively. Furthermore, it is assumed that if an electron resides in the ML, its spin $\hat{s} = (1/2) \sum_{\sigma \sigma'} \hat{d}^\dagger_\sigma \hat{n}_{\sigma \sigma'} \hat{d}_{\sigma'}$ [with $\sigma \equiv (\sigma^+, \sigma^0, \sigma^-)$ denoting the Pauli matrices] couples via ferromagnetic exchange interaction $J > 0$ to the core spin $\hat{S}_c$, so that the total spin of the molecule reads as $\hat{S} = \hat{S}_c + \hat{s}$. Finally, the ML is considered to be proximized by a SC lead [depicted in Fig. 1(b) as a shaded region], with the relevant coupling strength $\Gamma$.

In the large-gap limit, the effective Hamiltonian for such a SC-proximized molecule takes the form [34, 38–40]

$$\hat{H}_{\text{mol}} = \hat{H}_{\text{ML}} - J \hat{S}_c \cdot \hat{s} - \Gamma_S [\hat{d}_\uparrow \hat{d}_\downarrow + \text{h.c.}].$$ (1)

Next, the leads [with $q = \text{L(efl)}, R(ight)$] of the junction are described as reservoirs of spin-polarized, non-interacting itinerant electrons, $\hat{H}_{\text{leads}} = \sum_{k \sigma} \varepsilon_k \hat{c}^\dagger_{k\sigma} \hat{c}_{k\sigma}$. The operator $\hat{c}^\dagger_{k\sigma} \hat{c}_{k\sigma}$ creates (annihilates) a spin-$\sigma$ electron with energy $\varepsilon_k$ and momentum $k$ in the $q$th lead. Tunneling of electrons between leads and the molecule is captured by $\hat{H}_T = \sum_{k \sigma} \sqrt{\Gamma_\sigma} \varepsilon_k \hat{c}^\dagger_{k\sigma} \hat{d}_\sigma + \text{h.c.}$, where $\Gamma_\sigma$ denotes the spin-dependent hybridization function. In addition, a symmetric and flat conduction band within the range $[-W, W]$ is assumed in both leads, with the constant density of states $\rho = 1/(2W)$. Assuming the left-right coupling symmetry, the tunnel-coupling to two transport leads can be reduced at equilibrium to a single-channel problem [41] with a new effective hybridization $\Gamma_\sigma = \Gamma_\sigma^L + \Gamma_\sigma^R$. Since the primary focus of this Rapid Communication is the formation of spintronic fields, we consider here only the parallel relative orientation of spin moments in leads. In such a configuration, one finds $\Gamma_{\uparrow\downarrow} = (\Gamma_{\uparrow} + \Gamma_{\downarrow})/2$ with $\Gamma_{\uparrow}$ and $\Gamma_{\downarrow}$ being the spin-polarizing leads.

Spin-resolved transport properties of the device are characterized by the normalized spectral function of the ML, $A_\sigma(\omega) = -\Gamma_\sigma \text{Im} \{ G^R_\sigma(\omega) \}$, where $G^R_\sigma(\omega)$ is the Fourier transform of the retarded Green’s function $G^R_\sigma(\omega) = -i\theta(\omega) \langle \{ \hat{d}_\sigma(t), \hat{d}^\dagger_\sigma(0) \} \rangle$. This allows us to find the linear-response conductance between the two ferromagnets, $G = G_\uparrow + G_\downarrow$, with $G_\sigma = (e^2/h) \int d\omega [-\partial f(\omega)/\partial \omega] A_\sigma(\omega)$, where $f(\omega)$ is the Fermi function. The spectral function together with other averaged relevant quantities are obtained with the aid of the NRG method [36, 42]—see [43] for the NRG parameters used in calculations.

![Figure 2](image-url)

**Figure 2.** (a) Spin-quadrupole moment $\langle Q_{zz} \rangle$ for a molecule of spin $S = 3/2$ plotted as a function of tunnel coupling $\Gamma$ for $\Gamma_s = 0$ and several spin polarizations $p$ of the leads. (b) $\langle Q_{zz} \rangle$ shown for different values of $\Gamma_s$ indicated in (d) and $p = 0.5$. Panels (c) and (d) present the linear-response conductance $G$ (normalized to $G_0 = 2e^2/h$) and the spin polarization of transported electrons $\gamma$, respectively, which correspond to curves shown in (b). Other parameters: $U/W = 0.5$, $J/U = 10^{-3}$ and $T/U = 10^{-10}$. Quadrupolar spintronic field.—Generally, $\langle S_z \rangle \neq 0$ implies that a (large) spin is subject to a magnetic field, either real (externally applied) or spintronic one, while $\langle Q_{zz} \rangle \neq 0$ indicates the presence of an uniaxial magnetic anisotropy, regardless of its origin. Since the main focus is here on the quadrupolar field, we assume henceforth that the ML is electrically tuned to the particle-hole symmetry point ($\varepsilon = -U/2$), where the dipolar field disappears and $\langle S_z \rangle = 0$ [14]. In Fig. 2(a) we show for a model molecule with $S = 3/2$ (i.e., $S_z = 1$) how $\langle Q_{zz} \rangle$ evolves when shifting between the regimes of weak (1) and strong (3) tunnel coupling $\Gamma$. For $p = 0$, the SU(2) spin symmetry guarantees $\langle Q_{zz} \rangle = 0$. However, as soon as $p \neq 0$, the quadrupolar spintronic field ($D < 0$) becomes active and the ground-state doublet $| \pm S_z \rangle$ gets stabilized due to the onset of the energy barrier $\Delta E = (2S - 1)|D|$ for spin reversal, see Fig. 1(a). It can be seen that $\langle Q_{zz} \rangle$ saturates to two different values for $|D| > T$: $\langle Q_{zz} \rangle \rightarrow S(2S - 1)/3 = 1$ in the intermediate tunnel-coupling regime (2), and $\langle Q_{zz} \rangle \rightarrow S_z(2S_z - 1)/3 = 1/3$ in the strong one (3). The reduction of $\langle Q_{zz} \rangle$ in the latter case stems from the arrival of the Kondo correlations, which lead to screening of the ML spin $\hat{s}$ [44], and, thus, effective reduction of the...
total spin of the molecule and the unitary conductance through the device, see the solid line in Fig. 2(c). Note that for $S = 1$ considered in Ref. [19], $S_c = 1/2$ so that the molecule is never anisotropic in the Kondo regime. Conversely, for $S \geq 3/2$ the magnetic core of the molecule remains anisotropic in the Kondo regime $\langle \langle Q_{zz} \rangle \rangle$, leading to finite $\langle Q_{zz} \rangle$, clearly seen in Fig. 2(a). However, due to the screening of the ML spin, it is difficult to address and measure the spin state of the core by means of only transport spectroscopy of the ML.

The three regimes of tunnel-coupling visible in Fig. 2(a) persist for a wide range of spin polarizations $p$ of leads. Indeed, already for $p = 0.01$ a narrow range of intermediate couplings $\Gamma$ can be observed—meaning that a high spin polarization of a ferromagnet is not a necessity for generating spintronic anisotropy. For larger $p$ this region broadens quickly at the expense of $\langle \langle Q_{zz} \rangle \rangle$. The latter mechanism is, however, efficient mainly for very large spin polarization; see the line for $p = 0.99$ in Fig. 2(a).

Another hallmark feature of the quadrupolar field is the enhanced spin-filtering of electrons tunneling through the device, quantified by $\gamma = (G_\uparrow - G_\downarrow)/(G_\uparrow + G_\downarrow)$, see solid line in Fig. 2(d). It occurs due to suppression of electron-tunneling-induced spin-exchange processes involving the axial states $|\pm S_2\rangle$. Analyzing how $\gamma$ depends on $\Gamma$, one can identify crossovers between all three coupling regimes, with $\gamma \to 0$ indicating the onset of the Kondo effect (when the conductance reaches unitary limit in both spin channels).

**Critical point of the proximized molecule.**—The situation changes substantially if the molecule becomes proximized by a SC ($\Gamma_S \neq 0$), cf. solid and dashed curves in Figs. 2(b)-2(d). First of all, one observes that for a finite $\Gamma_S$ the quadrupolar field sets in already at a much weaker tunnel coupling $\Gamma$, *i.e.*, for values of $\Gamma$ few orders of magnitude smaller as compared to $\Gamma_S = 0$, see Fig. 2(b). Then, at some critical value of $\Gamma_S^* / U = 0.5005$, the three regimes in $\langle Q_{zz} \rangle (\Gamma)$ cease to be visible, and $\langle Q_{zz} \rangle \approx 1/3$ in full range of $\Gamma$. For yet larger $\Gamma_S$, $\Gamma_S > \Gamma_S^*$, only two distinctive regimes emerge: The first one (for small $\Gamma$), when the molecule is practically spin-isotropic with $\langle Q_{zz} \rangle = 0$, and the second regime (for large $\Gamma$), when anisotropy appears, but $\langle Q_{zz} \rangle$ does not exceed $1/3$.

To understand this behavior, let us point out that the SC-proximized molecule isolated from normal leads experiences a quantum phase transition at $\Gamma_S = (U + JS_c)/2$ [45], analogous to that predicted for single [34, 46] or double quantum dots [47, 48]. Then, the ground spin-multiplet changes from $S = 3/2$ with odd occupied ML, for $\Gamma_S < \Gamma_S^*$, to two spin-triplets ($S = 1$), with the even occupation of ML, for $\Gamma_S > \Gamma_S^*$—implying the reduction of the molecule’s spin to $S = S_c$. This phenomenon, known as Yu-Shiba-Rusinov (YSR) screening [29], explains the absence of the regime with $\langle Q_{zz} \rangle = 1$ in Fig. 2(b) and a suppression of the spin-filtering effect visible in Fig. 2(d) for $\Gamma_S / U = 0.51$. Importantly, note that this is not the Kondo screening, so that the strong tunnel-coupling regime for $\Gamma_S > \Gamma_S^*$ does not correspond to the Kondo regime anymore.

In the vicinity of the critical point, even very small $\Gamma$ allows for excitations between the two spin-multiplets. For $\Gamma_S = \Gamma_S^*$, this results in induced anisotropy in the full tunnel-coupling range [see double-dotted line in Fig. 2(b)], and a residual spin-polarized conductance in the weak tunnel-coupling regime [Figs. 2(c)-2(d)]. As long as $\Gamma_S < \Gamma_S^*$, a resonance appears in the conductance whenever $\Gamma$ becomes of the order of $\Gamma_S^* - \Gamma_S$, see Fig. 2(c). More precisely, this resonance starts forming when the broadening of energy levels becomes smaller than the excitation energy between the even and odd ML occupation states, $\Gamma \lesssim \Gamma_S^* - \Gamma_S$. Then, the system enters the Kondo regime, visible as an enhanced conductance. However, it quickly drops again with lowering $\Gamma$ due to exponential dependence of the Kondo temperature $T_K$ on $\Gamma$ and the fact that temperature $T$ used in Fig. 2, albeit finite, is very small. Thus, if $T > T_K$, the conductance becomes suppressed with decreasing $\Gamma$, which is a manifestation of the Coulomb blockade effect. Clearly, the Kondo peak does not arise for $\Gamma_S \geq \Gamma_S^*$, since after crossing the parity-changing transition the molecule is in the YSR-screened ground state.

This phase transition is sharp only in the limit $\Gamma \to 0$, and becomes a smeared crossover for finite $\Gamma$ [34]—observe the increasing width of the corresponding resonance in $G(\Gamma_S)$ at $\Gamma_S \approx \Gamma_S^*$ in Fig. 3(a). Moreover, $\Gamma_S^*$ gets diminished for larger $\Gamma$, which explains the suppression of $\gamma$ for $\Gamma_S$ equal to bare $\Gamma_S^* / U = 0.5005$ in the strong coupling regime, see the double-dotted line in Fig. 2(d).

Finally, providing complementary information to $G$, also $\langle Q_{zz} \rangle$ is the convenient quantity for describing the critical behavior of a system, see Fig. 3(b). It changes

![Figure 3](image-url)
1. Abruptly at the transition point for small \( \Gamma \), dropping from \( \langle Q_{zz} \rangle = 1 \) (characteristic of \( S = 3/2 \) and \( D < 0 \)) to \( \langle Q_{zz} \rangle = 1/3 \) (typical for \( S = 1 \) at \( \Gamma_S = \Gamma_S^* \)). For \( \Gamma_S > \Gamma_S^* \), \( \langle Q_{zz} \rangle \) can decrease yet further owing to the suppression of magnetic core anisotropy in the YSR-screened phase. Still, large \( \Gamma \) allows the molecular core to remain anisotropic even deep in the YSR phase, similar to the Kondo phase for \( \Gamma_S < \Gamma_S^* \), see the dot-dashed curve.

2. **Spectral signatures of the SC-proximity.**—In experiments, the quadrupolar spintronic field can be accessed indirectly via transport measurements of the spectral properties of a device [50]. Particularly, as mentioned above, this field leads to the occurrence of an excitation gap \( \Delta E \) between the ground-state doublet \( \pm S_z \) and the first-excited doublet \( \pm S_z \), see Fig. 1(a). This excitation manifests itself as a pronounced step in the spectral function \( \mathcal{A}(\omega) \), see Fig. 4(a). This step in \( \mathcal{A}(\omega) \) becomes indistinguishable because the ML is almost fully conducting, i.e., \( A_0 \equiv A(\omega = 0) \approx 2 \), with 2 signifying two spin transport channels. Moreover, for \( \Gamma_S > \Gamma_S^* \), the YSR screening becomes important and magnetic anisotropy is suppressed.

To investigate the evolution of the gap \( \Delta E(\Gamma_S) \) for a wide range of \( \Gamma \), we estimate \( \Delta E \) from the inflection point of the step in \( \mathcal{A}(\omega) \), as schematically illustrated in Fig. 4(a). The result is plotted as points in Fig. 4(b), with the relative height \( \Delta A \) of corresponding steps used in calculations [see Fig. 4(a)] shown in Fig. 4(c). One observes that \( \Delta E \)—and thus, the spintronic anisotropy—can be enhanced by a few orders of magnitude by proximizing the molecule with a SC. To corroborate this finding, in Fig. 4(b) we also present results for \( \Delta E \) (lines) derived with another method that does not resort to the analysis of spectral features. Specifically, in this theoretical approach, one seeks for some hypothetical component of magnetic anisotropy \( D > 0 \) one would need to add to compensate the quadrupolar spintronic field \( D < 0 \). It corresponds to numerically (by means of NRG) finding \( D \) from the condition \( \langle Q_{zz} \rangle(\tilde{D}) = 0 \) [49], and \( \Delta E = (2S - 1)\tilde{D} \). The results obtained with these two methods agree. In fact, with the latter approach one can address the large-\( \Gamma \) regime where electronic transport is dominated by Kondo fluctuations, so that the examination of the step in \( \mathcal{A}(\omega) \) becomes impossible, see Fig. 4(c).

The mechanism leading to the enhancement of spintronic anisotropy by proximity effect is associated with a strong renormalization of Coulomb correlations in the molecule. Strong coupling to a SC mixes the empty and doubly occupied states, increasing the charge fluctuations responsible for the development of \( \Delta E \). These fluctuations grow with increasing \( \Gamma \), resulting in \( \Gamma^2 \)-dependence of induced anisotropy, which is clearly visible in the weak and intermediate coupling regimes. However, when with augmenting \( \Gamma \) the Kondo effect sets in, the \( \Delta E \sim \Gamma^2 \) dependence is no longer valid and \( \Delta E \) starts decreasing with further increase of \( \Gamma \). Finally, the asymptotic behavior of \( \Delta E(\Gamma) \) for \( \Gamma/U \rightarrow 1 \) is the same regardless of \( \Gamma_S \), see Fig. 4(b), since tunnel broadening of all molecular states is so large that all states become strongly hybridized.

**Conclusions.**—We have shown that the proximity of a superconductor can enhance the spintronic anisotropy by orders of magnitude in the regime of weak and intermediate tunnel coupling to normal leads. This effect is particularly strong in the vicinity of parity-changing transition of the SC-proximized molecule. We have also discussed the spectral features, which can give access to detailed quantitative information about the spintronic anisotropy. Our work provides new means for manipulating properties of large-spin molecules and sheds light on unexplored spintronic aspects of the proximity-induced pairing.

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Note that all quantities and parameters are set in units of energy, that is, $\hbar = |e| = k_B = 1$.

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Supplemental Material for

Giant superconducting proximity effect on spintronic anisotropy

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This Supplemental Material contains further discussion related to the results presented in the main text. In order to provide direct insight into the physical mechanism underlying the enhancement of spintronic anisotropy in the vicinity of the charge parity transition of a molecule, the exact energies and eigenstates of the system in the limit of a molecule decoupled from the normal leads are analyzed in Sec. I. Next, we discuss the local spectral density of the relevant molecular level in the full range of energies (Sec. II), with the main focus on commenting the features irrelevant for the analysis of the spintronic anisotropy, and thus, not addressed in the main text. In Sec. III we describe in detail the complementary theoretical approach used for calculating the induced anisotropy, which does not resort to examining of the spectral features. Finally, we extend the analysis conducted in the main text to a few more general cases and show that the qualitative understanding of the results remains valid. Specifically, these cases include different values of the molecule spin (Sec. IV) and the analysis of the example of an intrinsically anisotropic molecule (Sec. V).

Within the Supplemental Material references are numbered as, e.g., Eq. (S-1) and Fig. S-1, whereas regular numbers, that is, Eq. (1) and Fig. 1, refer to the main article.

I. SUPERCONDUCTOR-PROXIMIZED MOLECULE SPECTRUM

In this section we analyze the eigenstates and eigenvalues of a molecule proximized by a superconducting electrode, ΓS > 0, yet decoupled from ferromagnetic leads, Γ = 0. Furthermore, we consider here a more general form of the Hamiltonian \( \hat{H}_{\text{mol}} \) for a superconductor-proximized molecule,

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Eq. (1), that involves also the effect of intrinsic uniaxial magnetic anisotropy of a molecule. This effect is captured by adding a new term $D_0 \hat{Q}_{zz}$ to $\hat{H}_{\text{mol}}$, with $D_0$ standing for the anisotropy constant and
\[
\hat{Q}_{zz} \equiv \hat{S}_z^2 - \mathcal{S}(\mathcal{S} + 1)/3
\] (S-1)
being the $z$-component of the spin quadrupole moment tensor. Recall from the main text that the total spin of the molecule, $\mathcal{S} = \hat{S}_c + \hat{s}$, is composed of the core spin $\hat{S}_c$ and the spin $\hat{s}$ of an electron residing in a molecular level (ML). As a result, the molecular Hamiltonian reads as
\[
\hat{H}_{\text{mol}} = \varepsilon \sum_{\sigma} \hat{n}_{\sigma} + U \hat{n}_{\uparrow} \hat{n}_{\downarrow} - J \hat{S}_c \cdot \hat{s} - \Gamma_S [\hat{d}_r^\dagger \hat{d}_r + \text{h.c.}] + D_0 \hat{S}_z^2.
\] (S-2)
Note that for conceptual simplicity of the further discussion, in the equation above we have dropped the constant term $C = -D_0 \mathcal{S}(\mathcal{S} + 1)/3$. In fact, the value of $C$ is a function of the total spin of the molecule, which depends also on its charge state. However, as long as only the two charge states of lowest energies are relevant for the physics of the system and the spin of the ground state is well-defined, i.e. at $T \ll J, U$, the only consequence of $C$ dependence on $\mathcal{S}$ is a small renormalization of $U$ (of the order of $D_0$). Thus, it seems justified to neglect the constant term under discussion.

The additional anisotropy term in Eq. (S-2) will prove helpful in qualitative understanding of the physics behind the transition between two different ground states which a molecule undergoes at some critical value $\Gamma_S^c$. As will be shown in Sec. V, its inclusion does not affect qualitatively the key results discussed in the main text.

A. Exact eigenstates and eigenvalues

First of all, note that, since ferromagnetic leads are assumed here to be decoupled from a molecule, the spintronic anisotropy does not occur in the following description.

Despite its complex form, Hamiltonian (S-2) can still be solved exactly. For this purpose, let us introduce the suitable basis of the Hilbert space consisting of product states $|\text{core} \rangle \otimes |\text{ML} \rangle$. Here, the state of magnetic core is determined by the $z$-component of its spin $\hat{S}_c$, $|\text{core} \rangle \equiv |S_{c,z} \rangle$. On the other hand, the state of the molecular orbital (ML) is described by its occupation, that is, $|\text{ML} \rangle \in \{0, \uparrow, \downarrow, 2\}$, with 0 (2) representing the unoccupied (doubly occupied) orbital and $\sigma = \uparrow, \downarrow$ corresponding to a case of the orbital being occupied by a single electron with spin $\sigma$. Next, it is easy to observe that the $z$-component of the total spin, $S_z$, and the parity of the ML occupation, $\alpha$ (defined as $\alpha = 1$ for even- and $\alpha = -1$ for odd-parity states), are conserved quantities, and thus, good quantum numbers to be used for labeling the eigenstates. Moreover, the states with different $\alpha$ are also always characterized by different $S_z$ (being integer for one parity value and half-integer for the other). In consequence, the parity index $\alpha$ can be omitted in the eigenstate label, so that the eigenstates of Hamiltonian (S-2) are denoted as $|S_z, r \rangle$, with $r$ indexing states within a given $S_z$ subspace. For $J > 0$, the possible values of $S_z$ for the ground-state spin multiplet lie in the range from $-S_c - 1/2$ to $S_c + 1/2$. As long as $|S_z| \leq S_c$, all the $S_z$ subspaces are two-dimensional. Below we first obtain eigenenergies and eigenstates for the situation of the even occupation of ML, and next for the ML occupied by a single electron.

a. The even occupation of ML (i.e. for $|\text{ML} \rangle \in \{0, 2\}$ and $S_z = S_{c,z}$)—In such a case, the ML spin is $s = 0$ and the term proportional to $J$ in Eq. (S-2) does not contribute. It basically means that states with different values of $S_z$ are not mixed, and the two-dimensional Hamiltonian matrix for a given $S_z$ can be easily diagonalized, which yields the eigenenergies
\[
E_{S_{z,a}, a} = \varepsilon + U/2 + D_0 S_z^2 + \sqrt{\Gamma_S^2 + (U/2 + \varepsilon)^2},
\] (S-3a)
\[
E_{S_{z,b}, b} = \varepsilon + U/2 + D_0 S_z^2 - \sqrt{\Gamma_S^2 + (U/2 + \varepsilon)^2},
\] (S-3b)
and the corresponding eigenstates
\begin{align}
|S_{z, a}\rangle &= \frac{1}{\sqrt{2}} \left( \sqrt{1 - \gamma_{\text{even}}} |S_z\rangle \otimes |0\rangle - \sqrt{1 + \gamma_{\text{even}}} |S_z\rangle \otimes |2\rangle \right), \\
|S_{z, b}\rangle &= \frac{1}{\sqrt{2}} \left( \sqrt{1 + \gamma_{\text{even}}} |S_z\rangle \otimes |0\rangle + \sqrt{1 - \gamma_{\text{even}}} |S_z\rangle \otimes |2\rangle \right),
\end{align}
(S-4a) \quad (S-4b)
with the coefficient \( \gamma_{\text{even}} \) defined as
\begin{equation}
\gamma_{\text{even}} = \frac{\varepsilon + U/2}{\sqrt{\Gamma_S^2 + (\varepsilon + U/2)^2}}.
\end{equation}
(S-5)

b. The odd occupation of ML (i.e. for \(|\text{ML}\rangle \in \{\uparrow, \downarrow\}\) and \(|S_z = S_{\text{c}, z}| = 1/2\)—First of all, a pair of states corresponding to maximal value of \(|S_z| = S_{\text{c}} + 1/2\), namely,
\begin{equation}
|S_z = \pm(S_{\text{c}} + 1/2), \pm\rangle = |\pm S_{\text{c}}\rangle \otimes |\uparrow\rangle
\end{equation}
(S-6)
are trivial eigenstates with eigenenergies
\begin{equation}
E_{S_z=\pm(S_{\text{c}}+1/2),\pm} = \varepsilon - JS_{\text{c}}/2 + D_0S_z^2.
\end{equation}
(S-7)
The reason to take the auxiliary index \( r = - \) will become clear towards the end of this section. The solution of \( H_{\text{mol}} \) in the remaining subspaces reads
\begin{align}
E_{S_z,+} &= \varepsilon + J(S_{\text{c}} + 1/2) + D_0S_z^2, \\
E_{S_z,-} &= \varepsilon + JS_{\text{c}}/2 + D_0S_z^2,
\end{align}
(S-8a) \quad (S-8b)
for eigenenergies, and
\begin{equation}
|S_{z, \pm}\rangle = \frac{1}{\sqrt{2}} \left( \sqrt{1 \mp \gamma_{\text{odd}}(S_z)} |S_z - 1/2\rangle \otimes |\uparrow\rangle \mp \sqrt{1 \pm \gamma_{\text{odd}}(S_z)} |S_z + 1/2\rangle \otimes |\downarrow\rangle \right),
\end{equation}
(S-9)
for eigenstates, with the coefficient \( \gamma_{\text{odd}}(S_z) \) given by
\begin{equation}
\gamma_{\text{odd}}(S_z) = \frac{S_z}{S_{\text{c}} + 1/2}.
\end{equation}
(S-10)
We note that the states denoted in Eq. (S-9) as \(|S_{z, \pm}\rangle\) correspond to components of the spin multiplet \( \mathcal{S} = S_{\text{c}} + 1/2 \), while those represented as \(|S_{z, +}\rangle\) constitute the spin multiplet \( \mathcal{S} = S_{\text{c}} - 1/2 \). Moreover, one can notice that Eq. (S-7) is formally equivalent to Eq. (S-8b)—justifying our choice of \( r = - \) in Eqs. (S-6)-(S-7)—which is expected because the spin multiplet \( \mathcal{S} = S_{\text{c}} + 1/2 \) should be split only via the term proportional to \( D_0 \). We also remind that eigenenergies \( E_{S_z,-} \) and \( E_{S_z,+} \), Eq. (S-8), should be understood as the ground-state energies for the spin multiplet \( \mathcal{S} = S_{\text{c}} + 1/2 \) (if \( J > 0 \)) and \( \mathcal{S} = S_{\text{c}} - 1/2 \) (if \( J < 0 \)), respectively. In the limit of non-magnetic leads \((p = 0)\) and for \( D_0 = 0 \), as considered in Fig. 2(a) in the main text, the \( SU(2) \) spin symmetry is recovered and the states in all respective spin multiplets are degenerate.

Finally, let us point out that the model of a molecule used in this work, and represented by the Hamiltonian \( H_{\text{mol}} \), Eq. (S-2), differs slightly from that used by Misiorny et al. in Ref. [1]. The difference concerns the way how the \textit{intrinsic} uniaxial magnetic anisotropy is included into the model. Specifically, here we assume that the \textit{total} spin \( \mathbf{\hat{S}} \) of the molecule is subject to magnetic anisotropy, while in Ref. [1] only the \textit{core} spin \( \mathbf{\hat{S}}_{\text{c}} \) is affected by magnetic anisotropy.

**B. Andreev bound states**

Let us begin with the case of \( \Gamma_{\mathbf{\hat{S}}} = 0 \) and a physically reasonable limit of \(|D_0| \ll |J| \ll U\). Then, for \(|\varepsilon + U/2| \ll U\), the ground state of a molecule corresponds to the ML occupied by a single
FIG. S-1. Energies of Andreev bound states $\Delta E_{a(b)}^{\text{ABS}}$ [see Eq. (S-12)] presented as a function of the coupling strength $\Gamma_S$ of the ML to a superconductor in the case of the ferromagnetic exchange coupling between spins of the ML and the magnetic core of a molecule. Parameters used: $S_c = 1$, $\delta/U = 0.1$, $J/U = 10^{-3}$ and $D_0 = 0$.

electron. As in the main text, we focus on the situation when the exchange coupling $J$ arising between the spin of such an electron and the core spin of the molecule is ferromagnetic ($J > 0$), which means that the ground spin multiplet is characterized by spin $S = S_c + 1/2$. The relevant excited charge multiplets correspond to empty or doubly occupied ML, with excitation energies of the order of $U/2 \pm \delta$, where $\delta = \varepsilon + U/2$ is the detuning from the particle-hole symmetry point—note $\delta = 0$ in the main text.

The situation becomes more complex for $\Gamma_S > 0$, as in such a case the empty and doubly occupied states of ML are hybridized due to the proximity of a superconductor, see Eq. (S-4). Nevertheless, the two charge multiplets with the even occupation of ML remain relevant excitations, forming so-called Andreev bound states. Specifically, at low temperature ($T \ll 2S_c|D_0|, J$), when only the ground-state spin doublet $|\pm(S_c + 1/2), -\rangle$ is occupied, energies of relevant excitations are defined as

$$\Delta E_{a(b)}^{\text{ABS}} = E_{S_c,a(b)} - E_{S_c+1/2,-} = E_{-S_c,a(b)} - E_{-(S_c+1)/2,-}, \quad (S-11)$$

gives

$$\Delta E_a^{\text{ABS}} = U/2 + JS_c/2 + |D_0|(S_c + 1/4) + \sqrt{\Gamma_S^2 + \delta^2} \quad \text{(for } J > 0\text{)}, \quad (S-12a)$$

$$\Delta E_b^{\text{ABS}} = U/2 + JS_c/2 + |D_0|(S_c + 1/4) - \sqrt{\Gamma_S^2 + \delta^2} \quad \text{(for } J > 0\text{)}. \quad (S-12b)$$

Note that there exist also other Andreev bound states, involving transitions between different excited states. However, they are insignificant for the transport properties analyzed here and in the main article, so that we do not discuss them here in detail.

In Fig. S-1 we illustrate how energies of Andreev bound states evolve as a function of the coupling $\Gamma_S$ for $\delta > 0$. At $\Gamma_S = 0$, the two energies correspond to the excitation energies to empty ($\Delta E_a^{\text{ABS}}$) or doubly occupied ($\Delta E_b^{\text{ABS}}$) states of ML, which are of the order of $U/2$, split by particle-hole asymmetry $\delta$. Additional spin-related contribution to the splitting occurs also due to the interaction of the ML spin with the magnetic core of the molecule, $JS_c/2$, as well as due to magnetic anisotropy, $|D_0|(S_c + 1/4)$. In the proximity of a superconductor, $\Gamma_S > 0$, the two channels of excitations no longer correspond to particle or hole processes. On the contrary, two Andreev bound states are formed, whose energies develop continuously from the values observed at $\Gamma_S = 0$, always increasing the splitting as $\Gamma_S$ grows. Note that the spin-related contribution to the splitting of the two channels does not depend on $\Gamma_S$.

Importantly, one can observe in Fig. S-1 that the excitation energy associated with the transition denoted by ‘b’ becomes equal to 0 when $\Gamma_S$ is tuned to some critical value $\Gamma_S^* = \Gamma_S^*$,

$$\Gamma_S^* = \sqrt{[U/2 + JS_c/2 + |D_0|(S_c + 1/4)]^2 - \delta^2} \quad \text{(for } J > 0\text{)}. \quad (S-13)$$
In fact, $\Gamma_S^*$ is the quantum phase transition point, as the negative excitation energy essentially means the change in the ground state—with the new phase for $\Gamma_S > \Gamma_S^*$ being the Yu-Shiba-Rusinov screened (evenly-occupied) ML [2].

In the case considered in the main text, that is, for an intrinsically spin-isotropic molecule ($D_0 = 0$) tuned to the particle-hole symmetry point ($\delta = 0$) and attached to ferromagnetic leads, the anisotropy constant $D_0$ in Eq. (S-13) should be replaced by the quadrupolar spintronic field $D$.

However, for parameters used there ($J/U = 10^{-3}$ and $S_c = 1$), the contribution to $\Gamma_S^*$ due to spintronic anisotropy is of the order of $|D|/U \ll 10^{-6}$, and thus, it can be neglected. Consequently, the estimated value of the relevant quantum phase transition point is

$$
\frac{\Gamma_S^*}{U} \approx (1 + JS_c/U)/2 = 0.5005.
$$

Finally, to complete the current discussion, we note that an analogous analysis can be conducted also for the case of the antiferromagnetic exchange coupling between the ML and core spins, $J < 0$. Then, the key difference with respect to the case of $J > 0$ discussed above is that the state acting as the ground-state spin doublet for a singly occupied ML is $|\pm(S_c - 1/2), +\rangle$. As a result, the excitation energies associated with Andreev bound states are

$$
\Delta E_{a(b)}^{ABS} \equiv E_{S_c,a(b)} - E_{S_c-1/2,+} = E_{-(S_c,a(b))} - E_{-(S_c-1/2),+},
$$

which gives

$$
\Delta E_a^{ABS} = U/2 + |J|S_c/2 - |D_0|(S_c - 1/4) + \sqrt{\Gamma_S^2 + \delta^2} \quad \text{(for $J < 0$)},
$$

$$
\Delta E_b^{ABS} = U/2 + |J|S_c/2 - |D_0|(S_c - 1/4) - \sqrt{\Gamma_S^2 + \delta^2} \quad \text{(for $J < 0$)},
$$

and thus, the critical value of $\Gamma_S^*$ takes the following form

$$
\Gamma_S^* = \sqrt{[U/2 + |J|S_c/2 - |D_0|(S_c - 1/4)]^2 - \delta^2} \quad \text{(for $J < 0$)}.
$$

**II. DISCUSSION OF THE SPECTRAL FUNCTION PROPERTIES IN FULL RANGE OF ENERGIES**

Figure S-2 presents the spectral functions of ML, $A(\omega)$, in the full range of energies $\omega$. Analogously as Fig. 4(a) in the main text, Fig. S-2 illustrates the evolution of the spectra for different $\Gamma_S$. However, in order to resolve better the high-energy features in the spectral function, here $A(\omega)$ is plotted on a logarithmic scale and without rescaling it by $A_0 \equiv A(\omega = 0)$. We also show an additional curve for $\Gamma_S/U = 0.51$, which is not considered in Fig. 4.

The first observation is that $A_0$ is very small for $\Gamma_S = 0$, with $A_0/U \approx 10^{-5}$. This effect stems from the fact that for the tunnel coupling under consideration, $\Gamma/U \approx 10^{-3}$, temperature used in calculations, $T/U = 10^{-10}$, is still larger than the Kondo temperature, $T > T_K$. Thus, the results obtained for such parameters at the particle-hole symmetry point ($\delta = 0$) actually correspond to the Coulomb blockade regime. Since the conductance $G$ is determined there by electron cotunneling processes and it scales as $G(\Gamma) \propto \Gamma^2$, it is small in the limit of the weak tunnel coupling (i.e., when $\Gamma$ is small). On the other hand, the conductance increases when the co-tunneling processes are enhanced which takes place when the excitation energies to the states with the even occupation of the ML diminish—as it happens when increasing $\Gamma_S$; see Sec. I.

As explained in the main text, there appears a step in the spectral function localized at $\omega = \Delta \epsilon = (2S - 1)|D|$, whose position becomes shifted towards higher energies as $\Gamma_S$ gets augmented. However, at the same time, the relative height of the step becomes diminished to such an extent that in the vicinity of the critical point, $\Gamma_S \approx \Gamma_S^*$, one cannot distinguish the step any longer; see the curve for $\Gamma_S/U = 0.5$ in Fig. S-2. Positions of the steps for different $\Gamma_S$ agree with the values of $\Delta \epsilon$ estimated from the compensation method described in Sec. III, and they are indicated in Fig. S-2 by the arrows. Note that even though the compensation method allows for calculating $\Delta \epsilon$ for arbitrary values of $\Gamma_S$, no particular spectral features appear in $A(\omega)$ for
Γ_S ≥ Γ_S^*, i.e., close to or within the Yu-Shiba-Rusinov–screened phase. For this reason, we do not present in Fig. 4 results for Γ_S ≥ Γ_S^*.

Furthermore, in the limit Γ_S ≪ Γ_S^*, the spectral function exhibits also a second step at energies |ω| ≈ J, marked by the finely dashed vertical line in Fig. S-2. This step arises naturally in the present model due to the ferromagnetic exchange coupling J, and it corresponds to transitions between the ground-state doublet |±(S_e + 1/2), −⟩ (belonging to the spin multiplet S = S_e + 1/2) and the doublet |±(S_e − 1/2), +⟩ of the excited multiplet of spin S = S_e − 1/2. For Γ_S ≥ Γ_S^*, this step becomes suppressed by charge fluctuations, which become very efficient close to the critical point Γ_S, as explained in the following.

At Γ_S = 0, charge fluctuations require very high energies, ω ≈ U/2, and correspond to the so-called Hubbard peak in the spectral density A(ω). However, due to the proximity of a superconductor, Γ_S > 0, the excitation energy associated with charge fluctuations is decreased, see Eq. (S-12b), until it reaches 0 at Γ_S = Γ_S^*, and next increases again after the new ground state has been established. This effect is visible in Fig. S-2, where the position of the Hubbard peak becomes shifted towards smaller energies with Γ_S approaching the critical point as ω ≈ ΔE_{Hub}^{ABS}(Γ_S), and it gets shifted back towards larger ω for Γ_S > Γ_S^*, as expected from Eq. (S-12b). Note, however, that due to the finite tunnel coupling Γ, the critical point is slightly shifted from Γ_S = Γ_S^* to a value smaller by ∼ Γ, cf. Fig. 3. This fact is the reason why the Hubbard peak for Γ_S = Γ_S^* in Fig. S-2 is not positioned at ω = 0, but approximately at ω/U = Γ/U = 10^{-3}. At the same time, the excitation energy in the second excitation channel follows ω ≈ |ΔE_{Hub}^{ABS}(Γ_S)|, increasing with Γ_S according to Eq. (S-12a). This can be observed in Fig. S-2 as the second Hubbard peak occurring for Γ_S > 0 at ω/U > 1/2.

### III. COMPENSATION METHOD FOR CALCULATION OF INDUCED ANISOTROPY

As mentioned in the introductory section of the main text, the effective Hamiltonian describing magnetic molecule has a general form

\[ \hat{H}_{\text{eff}} = B S_z + D Q_{zz} \]  

(S-18)

with B and D representing magnetic field and magnetic anisotropy, respectively. While assuming that B = 0, we focus here on analyzing the role played by finite D, which can be either of intrinsic (due to spin-orbit interaction [3–5]) or extrinsic (i.e., generated spintronically [1]) origin. As a
result, one can in general think of \( D \) as composed of two contributions, \( D = D_0 + D_{\text{spin}} \), with \( D_0 \) representing the intrinsic contribution [introduced in Eq. (S-2)] and \( D_{\text{spin}} \) standing for the spintronic contribution, which arises due to the proximity of ferromagnets. Note that in the main text we consider the case of an intrinsically spin-isotropic molecule \( (D_0 = 0) \), so that \( D = D_{\text{spin}} \). In real systems, \( D_0 \) is a property of a particular type of molecule and it can be either positive or negative, whereas \( D_{\text{spin}} < 0 \), since the second-order electron tunneling processes always decrease the energy of the ground state [1].

Importantly, in a system tuned to the particle-hole symmetry point \( (\delta = 0) \)—to avoid the occurrence of the spintronic dipolar field—\( D_{\text{spin}} \) can be adjusted in a limited range by changing the tunnel coupling to ferromagnetic leads or by varying the spin polarization of the leads. Thus, one can imagine that in a molecule with small ‘easy-plane’ magnetic anisotropy \( (D_0 > 0) \) that is attached to ferromagnets, it is in principle possible to compensate \( D_0 \) by tuning \( D_{\text{spin}} \), so that the molecule becomes effectively spin-isotropic. This situation corresponds to exact vanishing of the spin-quadrupole moment, Eq. (S-1), at all temperatures \( T \), \( \langle Q_{zz} \rangle(T) = 0 \). On the other hand, \( \langle Q_{zz} \rangle > 0 \) indicates the ‘easy-axis’ magnetic anisotropy \( (D < 0) \), and \( \langle Q_{zz} \rangle < 0 \) is a hallmark of the ‘easy-plane’ magnetic anisotropy \( (D > 0) \).

Actually, based on the discussion above, we come up with a particularly convenient approach allowing for theoretical analysis of the induced magnetic anisotropy \( D_{\text{spin}} \) for a given model. Specifically, we treat the intrinsic component of magnetic anisotropy—in this context denoted by \(-\overline{D}\)—as a parameter that can be changed in calculations to obtain \( \langle Q_{zz} \rangle(D) \). The opposite sign of \( D \) reflects its hypothetical character, in contrast to a clear physical meaning of \( D_0 \). Then, \( D_{\text{spin}} \) is equal to \( \overline{D} \) that satisfies the condition \( \langle Q_{zz} \rangle(D) = 0 \). Within such a theoretical framework, which in the main text we refer to as the compensation method, \( D_{\text{spin}} \) has a precise meaning for any set of model parameters, irrespective of its signatures in the local spectral density of ML.

The working principle of the method under discussion is illustrated in Fig. S-3. To begin with, we first calculate \( \langle Q_{zz} \rangle(D) \) in the presence of only normal, metallic and non-magnetic leads. This case is plotted in Fig. S-3 with black lines: a solid line for \( S = 3/2 \), as in the main text, and, for comparison, with a dashed line for \( S = 1 \). Clearly, for \( \overline{D} < 0 \) and \( |\overline{D}| \ll T \), \( \langle Q_{zz} \rangle(D) \), i.e., for large positive (‘easy-plane’) hypothetical \( D_0 \),

\[
\langle Q_{zz} \rangle = (S_z^{\text{min}})^2 - S(S + 1)/3 < 0 \quad \text{for } \overline{D} < 0 \text{ and } |\overline{D}| \ll T,
\]

where the minimal \( z \)-component of the molecular spin is \( S_z^{\text{min}} = 0 \) for integer and \( S_z^{\text{min}} = 1/2 \) for half-integer value of the spin. At \( \overline{D} = 0 \), the \( SU(2) \) spin symmetry guarantees \( \langle Q_{zz} \rangle = 0 \), which
FIG. S-4. Comparison of the spectral functions of ML, \( A(\omega) \), for a finite spintronic component \( D_{\text{spin}} \) to magnetic anisotropy (shown as solid lines; the same as in Fig. S-2) with those for \( p = 0 \) (thus, for \( D_{\text{spin}} = 0 \), but with \( D_0 = -\tilde{D} \); dashed lines). The arrows indicate \( \omega = D_0 \) for the corresponding curve. Dotted lines present the results for the spin-anisotropic case \((p = 0 \text{ and } D_0 = 0)\). Other parameters as in Fig. S-2.

is for finite \( T \) obtained smoothly with increasing \( \tilde{D} \). Furthermore, similar smooth increase is obtained for positive \( \tilde{D} \), until \( \langle Q_{zz} \rangle \) reaches for \( \tilde{D} \gg T \) its maximal value

\[
\langle Q_{zz} \rangle = S^2 - S(S + 1)/3 \quad \text{for } \tilde{D} > 0 \text{ and } \tilde{D} \ll T.
\]  

(S-20)

The situation changes substantially when the normal non-magnetic leads become replaced by ferromagnets. Then, the asymptotic values of \( \langle Q_{zz} \rangle(\tilde{D}) \) remain the same as above, but the zero is shifted to a finite negative value of \( \tilde{D} \). This shift basically corresponds to the cancellation of the ‘easy-plane’ magnetic anisotropy of a hypothetical molecule for a given \( p \), \( \Gamma \), \( J \), and other model parameters. In other words, the molecule becomes spin-isotropic due to the presence of a spintronic component of magnetic anisotropy, which implies that the quadrupolar exchange field \( D_{\text{spin}} \) is of the ‘easy-axis’ type \((i.e., D_{\text{spin}} < 0, \text{as expected})\), and its magnitude \( D_{\text{spin}} = \tilde{D} \) can be determined numerically from the condition \( \langle Q_{zz} \rangle(\tilde{D}) = 0 \).

It is worth emphasizing that the key advantage of the method for deriving \( D_{\text{spin}} \) described above is that \( D_{\text{spin}} \) can also be calculated in this way in the strong-coupling Kondo regime and in the Yu-Shiba-Rusinov phase, where the usual spectral signatures of the magnetic anisotropy are suppressed. Moreover, a direct comparison with the results obtained from the analysis of the relevant spectral function (see Fig. 4) shows that the compensation method provides in fact a very accurate tool for calculating \( D_{\text{spin}} \) also in the regime, where it can be effectively estimated from the ML spectral function. On the other hand, the main technical disadvantage of the method is that each point on the \( \langle Q_{zz} \rangle(\tilde{D}) \)-plot corresponds to a separate run of NRG calculation, and for very small \( \tilde{D} \) one needs to keep very low \( T \) and a large number of states at each iteration step in order to obtain reliable results. In practice, this inconvenience can be partially circumvented by iteratively increasing the sampling of \( \langle Q_{zz} \rangle(\tilde{D}) \) in the vicinity of the zero, which can significantly reduce the necessary number of points. Finally, note that a finite physical \( D_0 \) by definition only shifts the values of \( \tilde{D} \) by a constant, therefore, taking it into account do not introduce any additional complications.

In order to corroborate that the compensation method and the analysis of spectral features in transport indeed yield the same results for the spintronic magnetic anisotropy \( D_{\text{spin}} \), in Fig. S-4 we directly compare the relevant spectral densities of ML corresponding to three situations:

1. A spin-isotropic \((D_0 = 0)\) molecule attached to ferromagnetic leads characterized by the spin-polarization coefficient of \( p = 0.5 \) (solid lines); \( D = D_{\text{spin}} \) can be estimated as \( \tilde{D} \) using the compensation method.
(2) A spin-anisotropic molecule with $D_0 = \tilde{D}$ corresponding to the case (1) attached to non-magnetic leads (i.e., for $p = 0$), implying that $D_{\text{spin}} = 0$ and $D = D_0$ (dashed lines).

(3) The case of $p = 0$ and $D_0 = 0$ for comparison (dotted lines).

One can see that for $|\omega| \gg D$ the spectral functions $A(\omega)$ corresponding to (2) and (3) are indistinguishable, while for (1) they are slightly reduced for $|\omega|/U < 1/10$. This is a consequence of the proximity of ferromagnetic lead.

See Sec. II for discussion of the spectral features at $|\omega| \gg D$. For weak enough $\Gamma_S$, curves corresponding to cases (1) and (2) exhibit a step at $\omega \approx \Delta \mathcal{E} = (2S - 1)|D|$ and a plateau for lower energies—both being characteristic signature of the presence of magnetic anisotropy. In all cases (i.e., also the step for large $\Gamma_S$ is absent) curves for (1) and (2) differ only by a small rescaling factor due to finite $p$ in (1). This is yet another prove that the compensation method for determining $D_p$ allows for calculation of the physically relevant quantity and does not lead to non-physical predictions concerning spectra, even in the regimes where the spectral signatures indicating magnetic anisotropy are difficult to recognize.

IV. DIFFERENT VALUES OF MAGNETIC MOLECULE SPIN

In the main text we considered a hypothetical molecule characterized by the total spin of $S = 3/2$. In the case of the ferromagnetic exchange coupling $J$ within the molecule, see Eq. (S-2), and the ML occupied by a single electron, such magnitude of the total spin is observed for the spin of the magnetic core of the molecule equal to $S_c = 1$. In the present section we discuss the influence of the value of $S_c$ on the results.

First of all, the effect of magnetic anisotropy reveals itself only if $S \geq 1$, and thus, we consider only $S_c \geq 1/2$. Furthermore, $S = S_c + 1/2$ is not valid in all the coupling regimes. This has been discussed in the main text in the context of Fig. 2(b) and is clearly visible also in Fig. S-5(a), where the expectation value $(Q_{zz})$ as a function of the tunnel-coupling $\Gamma$ is presented for different values of $S_c$ and $\Gamma_S$. Let us first discuss the set of curves for $\Gamma_S = 0$. Evidently, one observes $(Q_{zz}) \rightarrow 0$ in the limit where the tunnel-coupling is small and it increases to its maximal value, $(Q_{zz}) = S^2 - S(S + 1)/3$, in the intermediate tunnel-coupling regime, where $S_c = \pm S$ and $S = S_c + 1/2$. By further augmenting $\Gamma$, the onset of the Kondo screening in the strong tunnel-coupling regime becomes eventually unavoidable, and it leads to reduction of the effective spin of the molecule to $S = S_c$. Moreover, as discussed in Sec. I, the increase of $\Gamma_S$ drives a transition to the evenly-occupied ML state at $\Gamma_S = \Gamma_S^* \equiv (U + S_cJ)/2$, see Eq. (S-14), which also results in $S = S_c$. For this reason, it becomes clear that $S_c = 1$ is the smallest possible value of $S_c$ that guarantees that the effects associated with finite magnetic anisotropy should be expected to arise in all the regimes of $\Gamma$.

In general, as one can see in Fig. S-5, the behavior of the system with increasing $\Gamma_S$ toward the critical value $\Gamma_S^*$ for different values of $S_c$ is quantitatively the same as for the case of $S_c = 1$ studied in the main text. Still, for a given value of $\Gamma_S$ close to $\Gamma_S^*$, one can observe some large quantitative differences, visible especially in the position of the resonance in the conductance that is associated with the excitation energy between the oddly and evenly occupied-ML states, see in particular curves for $\Gamma_S/U = 0.5$ in Fig. S-5(b). The shift of this position stems from the dependence of the critical point $\Gamma_S^*$ on $S_c$, see Eq. (S-14). Finally, worthy of note is that the spin polarization of transported electrons, $\gamma$, does not depend significantly on $S_c$ in the limit of $\Gamma \rightarrow 0$, except at the critical point $\Gamma_S = \Gamma_S^*$, which is illustrated in Fig. S-5(c).

The most important point concerning the spin of the molecule is the lack of increase of the energy barrier with increasing $S$. Even though one could naively expect the relation $\Delta \mathcal{E} = (2S - 1)|D|$ to hold at least for relatively small $S$, in Fig. S-6(a) and (c) it is clearly visible that such a behavior is very weak. On the contrary, the energy barrier seems to be hardly affected by changing $S$, cf. Fig. S-9 in Supplementary Information of Ref. [1]. Moreover, in the vicinity of the critical point, where the superconductor proximity boosts the induced anisotropy most effectively, the energy barrier $\Delta \mathcal{E}$ appears to be decreasing for larger values of $S$, see gray curves in Fig. S-6(c). This effect is caused by the fact that the distance from the critical point, $\delta \Gamma_S \equiv \Gamma_S^* - \Gamma_S$, is decisive for
obtaining the highest anisotropy. Since $\Gamma_S^* \propto S_c$, see Eq. (S-14), the largest possible value of the molecular spin $S$ is not necessarily desired for applications, unless arbitrarily large coupling strengths $\Gamma_S$ are within reach. A similar conclusion can be drawn from the analysis of Fig. S-6(b), presenting the height of the step in the spectral functions $A(\omega)$ for different values of $S_c$. Clearly, it is the highest for $S_c = 1/2$ and diminishes with increasing $S_c$, which means that large $S_c$ is not necessarily the right choice for experiment or applications.
FIG. S-6. Extension of Fig. 4 to different values of the magnetic-core spin of the molecule, $S_c$. (a) Spectral function $A(\omega)$ [scaled by $A_0 \equiv A(\omega = 0)$] shown as a function of energy $\omega$ for selected values of $\Gamma_S$ and $\Gamma/U = 10^{-3}$. All spectral functions are symmetric with respect to $\omega = 0$. (b) The height $\Delta A/A_0$ of the step in $A(\omega)/A_0$ used for estimating $\Delta E$ in (c) plotted as a function of $\Gamma$. (c) Evolution of the excitation gap $\Delta E$ with $\Gamma$ (note a log-log scale). All other parameters as in Fig. 4.

V. RESULTS FOR FINITE INTRINSIC ANISOTROPY

In this final section we address the effects occurring due to the presence of finite intrinsic magnetic anisotropy ($D_0 \neq 0$), see Eq. (S-2). Specifically, our goal is to analyze here additional effects that originate from the interplay between the intrinsic ($D_0$) and spintronic ($D_{\text{spin}}$) components of magnetic anisotropy. We note that the key results presented in the main text remain qualitatively valid also for finite $D_0$. Our discussion we begin with the analysis of the case when there is no superconductor proximizing the molecule ($\Gamma_S = 0$), and in the next step we also take into consideration the superconductor-proximity effects.

A. Influence of intrinsic magnetic anisotropy ($D_0 \neq 0$) for $\Gamma_S = 0$

The occurrence of magnetic anisotropy in a large-spin molecule manifests clearly in the dependence of the expected value of the $z$-component of the spin-quadrupole moment tensor $\langle Q_{zz} \rangle$, Eq. (S-1), on the tunnel-coupling strength $\Gamma$, as presented in Fig. S-7(a). One can see there that, unlike for $D_0 = 0$, in the weak tunnel-coupling limit $\langle Q_{zz} \rangle > 0$ for $D_0 < 0$, while $\langle Q_{zz} \rangle < 0$ for $D_0 > 0$. In the case of intrinsic magnetic anisotropy of the ‘easy-axis’ type ($D_0 < 0$)—i.e., when
the ground-state spin-doublet $|\pm(S_z + 1/2), -\rangle$ is energetically preferable, see Sec. I—one expects that the anisotropic regime should persist also for small tunnel-couplings if only $\Delta E \ll T$ with no other qualitative consequences, which can be observed in Fig. S-7. On the contrary, for $D_0 > 0$ the competition between the intrinsic and spintronic components of magnetic anisotropy becomes apparent. This competition arises as a consequence of the fact that $D_{\text{spin}} < 0$, which basically means that the sign of the total effective magnetic anisotropy constant $D = D_0 + D_{\text{spin}}$ can be tunned by changing $\Gamma$. We note that for a half-integer total spin of a molecule $S$ (as considered here), positive $D$ implies that the two-fold degenerate ground state of a minimal $z$-component of spin, $|\pm 1/2, -\rangle$, is favored. Such a doublet is in fact spin-isotropic, that is, there is no energy barrier (in the absence of magnetic field) for switching between the states of the doublet. As a result, the shrinkage or even complete suppression [see the curve for $D_0/U = 10^{-4}$ in Fig. S-7(a)] of the anisotropic regime becomes visible whenever $D_0 > |D_{\text{spin}}|$. Moreover, if $D_0 \gg |D_{\text{spin}}|$, the Kondo spin-exchange processes within the ground-state doublet get enhanced, which leads to an increase of the Kondo temperature $T_K$. This behavior can be observed as a shift of the critical $\Gamma$ where the strong-coupling (Kondo) regime sets in towards smaller values, which is clearly visible in the dependence of the linear response conductance $G$ (normalized to $G_0 \equiv 2e^2/h$) as a function of $\Gamma$. Other parameters as in Fig. 2.

In principle, the total anisotropy $D$ can be predicted assuming that the dependence of the quadrupolar spintronic exchange field $D_{\text{spin}}$ on the intrinsic magnetic anisotropy $D_0$ is negligible, i.e., the induced anisotropy does not depend on the intrinsic one. Figure S-8(a) presents how...
FIG. S-8. Generalization of Fig. 4(b) for $\Gamma_S = 0$ to different values of intrinsic magnetic anisotropy of the molecule, $D_0$. All other parameters as in Fig. 4.

the position of the step in the spectral density of ML corresponding to spin-reversal excitations depends on the tunnel-coupling strength $\Gamma$. Clearly, for small $\Gamma$, when $D_p \sim \Gamma^2 \ll D_0$, the value of $|D| = \Delta E/(2S - 1)$ is in practice entirely determined by $D_0$. Then, for $D_0 < 0$, the estimated value of $D$ increases continuously and practically merges with $D_0 = 0$ curve, if only $|D_0| \ll \Delta E(D_0 = 0)$. For larger values of $|D_0|$, the influence of intrinsic magnetic anisotropy is visible as an additional increase of the energy barrier $\Delta E$; see curve for $D_0/U = 10^{-6}$.

The situation is qualitatively different for $D_0 > 0$, when the intrinsic ($D_0$) and spintronic ($D_{\text{spin}}$) contributions to magnetic anisotropy have the opposite sign. For small values of $\Gamma$, that is, if $\Gamma < \Gamma^*$ with the critical value $\Gamma^*$ defined via the relation $D_{\text{spin}}(\Gamma^*) = -D_0$, the behavior of the system is determined by $D_0$. Then, as discussed above, the ground-state doublet is $|\pm 1/2, -\rangle$, and the excitation energy $\Delta E$ corresponds to the leading transitions to the excited doublet $|\pm 3/2, -\rangle$. As the tunnel coupling $\Gamma$ grows and approaches its critical value $\Gamma^*$, this excitation energy gets diminished, and then completely suppressed at $\Gamma = \Gamma^*$, which means that the molecule becomes effectively spin-isotropic, with all states $|S_z, -\rangle$ (for $S_z = -S_c - 1/2, -S_c + 1/2, \ldots, S_c - 1/2, S_c + 1/2$) being degenerate at this point. Then, for even larger $\Gamma$, $\Gamma > \Gamma^*$, the spintronic contribution $D_{\text{spin}}$ starts dominating over the intrinsic component $D_0$ of magnetic anisotropy, and the spin doublet $|\pm (S_c + 1/2), -\rangle$ (or to be precise, $|\pm 3/2, -\rangle$ for $S_c = 1$) becomes restored as the ground state, with the energy barrier $\Delta E$ growing quadratically with $\Gamma$.

**B. Superconducting proximity effect ($\Gamma_S \neq 0$) for finite $D_0$**

The superconducting proximity effects for finite $D_0$ can be well understood by assuming that also for finite $\Gamma_S$ the induced anisotropy $D_{\text{spin}}$ is independent of $D_0$. In particular, the shift of the anisotropic regime towards smaller values of the tunnel-coupling $\Gamma$, visible in Fig. 2(b), still takes place for finite $D_0$, retaining the properties discussed in Sec. VA. Similar behavior is also reflected in the spectral properties, as illustrated in Fig. S-9. As expected, for all considered values of $\Gamma_S$, the position of the characteristic step in the ML spectral function remains fixed for $D_0 < 0$ in the weak tunnel-coupling limit and tends to shift only when $\Gamma$ is in the intermediate regime, where $|D_{\text{spin}}| \gtrsim |D_0|$. For $D_0 > 0$ the situation is similar, although for some critical value of the tunnel-coupling strength $\Gamma = \Gamma^*$ one gets $D_{\text{spin}} = -D_0$, so that the magnetic anisotropy is effectively removed and the step in the spectral function vanishes. Moreover, it is clear that due to the enhancement of $D_{\text{spin}}$ by orders of magnitude when a molecule is subject to the presence of a superconductor ($\Gamma_S \neq 0$), the condition $D_{\text{spin}} = -D_0$ can be fulfilled already at much smaller $\Gamma$. This corroborates the conclusion, that the induced spintronomically magnetic anisotropy $D_{\text{spin}}$ is in essence independent of $D_0$ and the separate analysis of the spintronic contribution conducted...
FIG. S-9. Analogous to Fig. 4(b), but now plotted also for selected values of the intrinsic magnetic anisotropy constant $D_0$. Note that fine-dashed lines serve as reference lines, as they correspond to the results for $D_0 = 0$ shown in Fig. 4(b). All other parameters as in Fig. 4.

in the main article remains valid also for spin-anisotropic molecules, with the values of the total magnetic anisotropy $D$ being shifted by $D_0$.

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