Indirect Spin-Readout of Rare-Earth-Based Single-Molecule Magnet with Scanning Tunneling Microscopy

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Rare-earth based single-molecule magnets are promising candidates for magnetic information storage including qubits as their large magnetic moments are carried by localized $4f$ electrons. This shielding from the environment in turn hampers a direct electronic access to the magnetic moment. Here, we present the indirect readout of the Dy moment in Bis(phthalocyaninato)dysprosium ($DyPc_2$) molecules on Au(111) using milli-Kelvin scanning tunneling microscopy. Because of an unpaired electron on the exposed Pc ligand, the molecules show a Kondo resonance that is, however, split by the ferromagnetic exchange interaction between the unpaired electron and the Dy angular momentum. Using spin-polarized scanning tunneling spectroscopy, we read out the Dy magnetic moment as a function of the applied magnetic field, exploiting the spin polarization of the exchange-split Kondo state.

Rare-earth-based single-molecule magnets (RE-SMMs) have been investigated extensively due to the potential application in data storage and quantum computing [1–12]. In RE-SMMs the strong spin-orbit coupling and the localization of the $4f$ orbitals prevents quenching of the $4f$ orbital angular momentum by the ligand field resulting in a favorable combination of large magnetic moments and large zero field splitting. Often, this effectively isolates the magnetic ground state(s) from excited states [7,13], rendering the system ideal to store and process information. A side effect of the rather localized $4f$ orbitals is further, that they are shielded from external perturbations promoting magnetic stability, i.e., long relaxation times T_1, as well as coherence times T_2. Magnetic bistability of long T_1 has been demonstrated in RE-SMMs for classical bits even above liquid nitrogen temperature [10]. Similarly, RE-SMMs including their nuclear spin were used as quantum bits (qubits) that can be electrically read and manipulated [1,5,7,8]. Practical coherence times T_2 have been achieved. However, the isolation of the $4f$ moment also brings an inherent difficulty. The localized $4f$ electrons hardly participate in electronic transport through the molecule, which has hindered the readout of the $4f$ moment in RE-SMMs using scanning tunneling microscopy (STM) [14].

We report on an indirect readout of the spin state of the Dy moment in Bis(phthalocyaninato)dysprosium ($DyPc_2$) double-decker molecules using spin-polarized STM (SP-STM) at around 50 mK. We find that the magnetic moment of central Dy^{3+} ion is ferromagnetically coupled to the singly occupied molecule orbital (SOMO) of the phthalocyanine (Pc) ligand in analogy to previous works in break junctions including $TbPc_2$ [1]. The Kondo effect in $TbPc_2$ and other rare-earth double deckers on the ligand is well established by the seminal work of Komeda and others [14–16]. With the high energy resolution of our instrument [17] we observed a splitting of Kondo peak of the unpaired electron without application of a magnetic field by the exchange coupling between the unpaired electron and the Dy momentum. Reading out the spin polarization of the exchange-split Kondo state as a function of the applied magnetic field allows us to determine the Dy moment indirectly. We demonstrate that the Kramers ground state doublet of Dy shows a smooth transition in the spin polarization rather than a abrupt switch. This is explained on the basis of the avoided level crossing of the lowest energy states and provides design rules for qubits based on RE-SMMs in contact to conductors.

The measurements were performed in a home-built ultrahigh vacuum (UHV) STM operating at a base temperature of about 25 mK with an external magnetic field up to 7 T oriented normal to the surface plane [17]. Spin-polarized STM tips were prepared by coating a W tip with
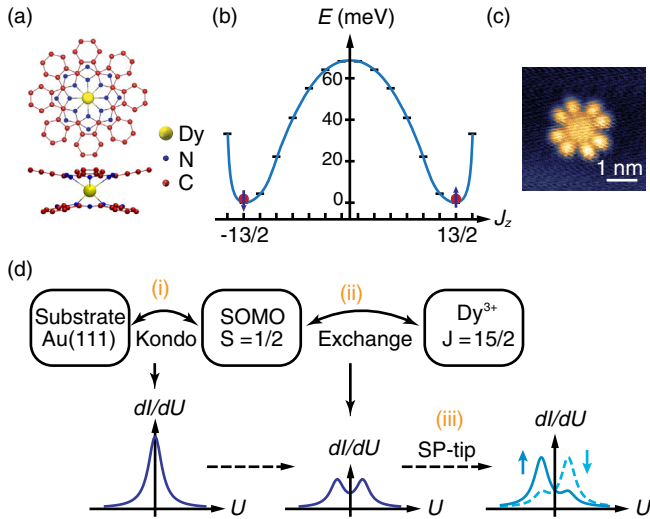
FIG. 1. (a) Top and side views of a free DyPc$_2$ molecule of D_{4d} symmetry. H atoms are omitted for clarity. (b) Zero field splitting of the magnetic states of the central Dy^{3+} ion. The energies and wave functions of the ground state multiplets are adopted from Ref. [24]. (c) STM topography of a DyPc$_2$ molecule on Au(111) ($U = 0.1$ V, $I = 5$ pA). (d) Basic idea to access the Dy magnetic moment: (i) The SOMO on the Pc ligand couples to the conduction electrons of the substrate causing a Kondo resonance. (ii) Magnetic exchange coupling between the SOMO and the Dy moment J splits the Kondo peak at zero magnetic field. (iii) With a spin-polarized tip, the magnetic moment of dysprosium can be read out indirectly.

FIG. 2. (a) Exchange splitting of the Kondo resonance as evidenced by local dI/dU spectra recorded at positions as indicated in the inset ($U = 1$ V, $I = 10$ pA). (b) Magnetic field dependence of the splitting of the Kondo resonance recorded on the lobes of the upper Pc ring. The spectra (colored dots) were measured using a lock-in modulation of $U_{mod} = 80$ μV and feedback conditions of $U = 5$ mV, $I = 10$ pA. Solid colored lines represent the fits to the data.

antiferromagnetic Cr. An out-of-plane spin polarization of the tip was achieved by adjusting the Cr thickness, and was cross-checked by imaging of Co/Cu(111) samples [18,19]. DyPc$_2$ molecules (see Supplemental Material [20] for synthetic method [21–23]) were deposited onto a clean Au(111) substrate held at room temperature from a Knudsen cell at 400 °C.

DyPc$_2$ consists of a central Dy^{3+} ion sandwiched by two Pc rings. The two Pc ligands are skewed against each other by 45° and adopt a bent configuration with respect to each other [see Fig. 1(a)] [23,24]. In the ideal case, the central ion possesses a D_{4d} symmetry and exhibits the electron configuration of [Xe]$4f^9$ [13,14]. According to Hund's rules, the Dy^{3+} multiplet is characterized by an angular momentum $L = 5$, a spin $S = 5/2$ and a total angular momentum $J = L + S = 15/2$, i.e., Dy^{3+} is a Kramers system. As a consequence, the zero field splitting results in Kramers doublets [13,24] as depicted in Fig. 1(b), that is, the $J_z = \pm 13/2$ ground states are separated from each other by an energy barrier of roughly 68 meV. Note that in the D_{4d} symmetry, J_z remains a good quantum number; i.e., the eigenstates are pure in J_z. This makes the DyPc$_2$ a promising SMM [13,24,25]. We note that the D_{4d} symmetry of the molecule adsorbed on Au(111) is broken reducing it to a C_4 symmetry allowing additional Stevens operators [26–28] in the zero field splitting, which will be
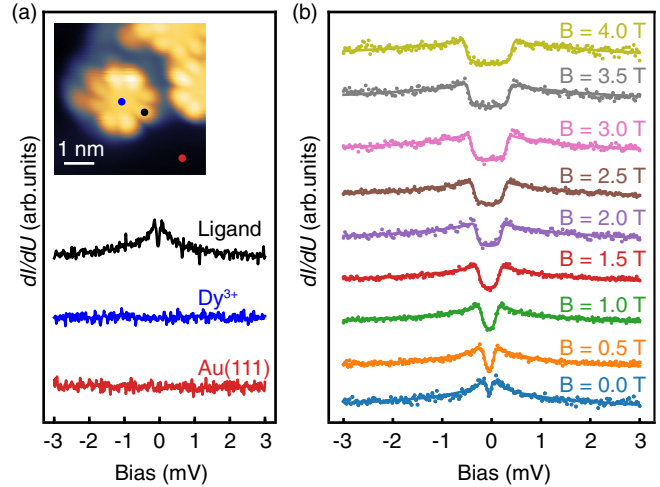
discussed later. In the STM, an isolated DyPc$_2$ molecule on Au(111) shows up as eight bright lobes corresponding to the SOMO of the upper Pc ligand [see Fig. 1(c)] on which we measured a Kondo resonance, i.e., the resonance is located on the ligand rather than the Dy center [14] [see Fig. 2(a) and Supplemental Material [20]].

Figure 1(d) shows the basic idea to access the Dy magnetic moment by STM. It is similar to the method introduced by Vincent et al. [1]. (i) When the molecules are deposited on the Au(111) surface, the coupling of the local spin of SOMO to the conduction electrons of substrate makes it possible for STM to observe the SOMO via a Kondo resonance, similar to other double-decker Pc molecules [14]. In contrast to the method of Vincent et al., no external gate is necessary but the Fermi energy of Au(111) with respect to the molecular orbitals ensures single occupancy. (ii) If the SOMO spin $1/2$ (S) exhibits an exchange interaction with the Dy magnetic moment J, the Kondo resonance is split by the exchange field, as also has been observed for TbPc$_2$ [7]. (iii) With a spin-polarized tip, the direction of polarization of the split Kondo peaks can be measured, allowing us to indirectly determine the Dy magnetic moment J. The split Kondo peaks are expected to be fully spin polarized [29] and thus, their intensity depends on the relative orientation of the tip and SOMO magnetic moment.

As an initial step, the differential conductance dI/dU on the DyPc$_2$ molecules was measured with unpolarized W tips at ≈ 50 mK and zero magnetic field as shown in Fig. 2(a). While the spectrum is essentially flat in the region

around the Fermi energy when recorded on the Au(111) substrate (red) or the center of the molecule (blue), i.e., above the Dy ion, a clearly split Kondo resonance (black) can be observed on the lobes of the upper Pc ligand. Since no magnetic field was applied, the splitting must originate from the exchange interaction between the SOMO spin 1/2 and the Dy magnetic moment. The Kondo resonance is strictly localized on the lobes of the upper Pc ring representing the SOMO (see Supplemental Material [20]).

To further elaborate on the split Kondo peaks, we recorded tunneling spectra as a function of an external magnetic field B applied perpendicular to the Au(111) surface. As can be seen from Fig. 2(b), the splitting gradually increases upon the increase of B, which is in line with the expectation. Note that a few hundred milli-Tesla are already enough to practically saturate the Dy Brillouin function at 50 mK (see also discussion, below). We thus expect a Dy moment aligned with the external magnetic field. In this case, the exchange field to the Dy ion and the external field add up to act on the SOMO spin 1/2. The width of the splitting linearly increases with magnetic field. For a quantitative analysis, we globally fit the experimental spectra with a model proposed in Ref. [30], which describes the detailed shape of the tunneling spectra using third-order perturbation theory. We treated the SOMO as a spin 1/2 system and the exchange interaction to the Dy ion as an effective exchange field B_{ex} [1]. The fits are presented in Fig. 2(b) as solid lines and reproduce all details of the experimental spectra. The global fit yields a ferromagnetic exchange field $B_{ex} = (422 \pm 20)$ mT, a Landé factor $g_s = (1.85 \pm 0.02)$ and an effective temperature $T_{eff} = (206 \pm 10)$ mK. The small deviation of the Landé factor from the ideal value $g = 2$ is expected for a delocalized spin 1/2. The fitted effective temperature of roughly 200 mK corresponds to the experimental broadening due to the finite lock-in modulation.

With these results, we proceed to the indirect readout of the Dy magnetic moment J using spin-polarized STM as depicted in Fig. 1(d). In essence, the splitting of the Kondo peak is related to the effective magnetic field that the unpaired electron on the SOMO feels, i.e., the sum of the external field and the exchange field. The two peaks of the split Kondo resonance are fully spin polarized [29], allowing us to determine the relative orientation of the spins of the unpaired electron and the tip. For instance, using a tip with a spin polarization along the positive z axis and an applied magnetic field also along the positive z axis, the state with $S_z = +1/2$ will be higher in energy and accordingly the peak above the Fermi level (i.e., at positive bias) will be higher in intensity than the one below. When the direction of the applied magnetic field is reversed, the opposite spin orientation with $S_z = -1/2$ will be higher in energy and the intensities of the two peaks swap roles. Thus, the asymmetry of the two peaks directly shows the orientation of the SOMO spin and indirectly

allows the readout of the Dy magnetic moment via the exchange interaction.

Figures 3(a) and 3(b) show a series of tunneling spectra of the split Kondo resonance measured at different magnetic fields taken with a Cr-coated, antiferromagnetic tip. For all applied fields, a splitting of the Kondo peak was observed, i.e., the ferromagnetic exchange field to the Dy ion is never compensated by the external field. Instead, the Dy moment seems to be reversed together with the unpaired electron near zero field. We observe a vanishing spin polarization of the Kondo peaks at about -10 mT. This small offset from zero magnetic field can be due to the coercivity of our superconducting magnet and/or to a small magnetic dipole field of the antiferromagnetic tip [31]. To extract the asymmetry of the Kondo peaks from the spectra, we used the same model as for the unpolarized tip [see Fig. 2(b)] with the addition of a fixed spin polarization of the Cr tip normal to the surface. The resulting asymmetry normalized to saturation is shown in Fig. 3(c) including the σ error of the fit.

To analyze the data in detail, we compare the experimental results with different models. First, we assume that the Dy moment is stable in time and only reverses when the sign of the external field reverses. In this scenario, the unpaired electron feels the Zeeman energy of the external field B and the exchange field B_{ex} of Dy. This scenario can be calculated based on the partition function Z of a spin 1/2 in the effective field, resulting in the Brillouin function offset by the exchange field:

$$\langle \hat{S}_z \rangle = A \cdot \tanh\left(\frac{g_s \mu_B (B + B_{ex})}{2k_B T}\right), \quad (1)$$

where μ_B is the Bohr magneton, T is the temperature, and k_B is the Boltzmann constant. The curve [blue line in Fig. 3(c)] does not fit the experimental data and the scenario can be safely excluded.

Next, we consider a model based on the partition function of the four lowest energy states of the coupled system while neglecting the off-diagonal Stevens operators (for details see Supplemental Material [20]). As the zero field splitting in the Dy system is much larger than the temperature, it is reasonable to consider only the four lowest states of the coupled system, i.e., $S_z = \pm 1/2$ coupled to $J_z = \pm 13/2$. This scenario leads to

$$\langle \hat{S}_z \rangle = \frac{1}{Z} \left(+\frac{1}{2}e^{-\beta\epsilon_1} - \frac{1}{2}e^{-\beta\epsilon_2} + \frac{1}{2}e^{-\beta\epsilon_3} - \frac{1}{2}e^{-\beta\epsilon_4} \right), \quad (2)$$

where $\beta = 1/k_B T$, $\epsilon_{1,2,3,4}$ correspond to the energy of the four lowest states. This scenario is represented in Fig. 3(c) with a green line. It nicely reproduces most of the experiment features, except the tiny overshoot at around ± 80 mT [see inset of Fig. 3(c)] which will be discussed later in the third model. This demonstrates that not only the unpaired
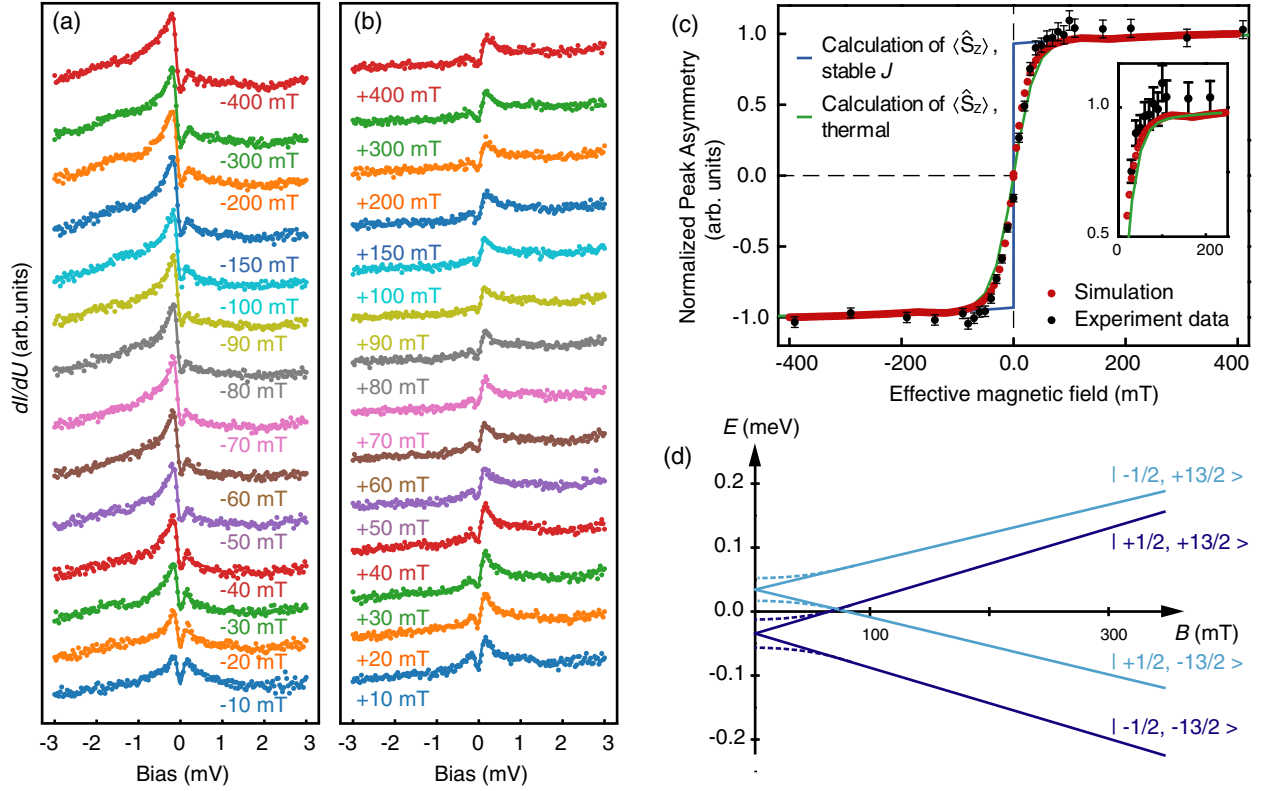
FIG. 3. (a)–(b) Spin-polarized scanning tunneling spectra of $DyPc_2$ as a function of applied magnetic field. The colored dots represent the experimental data ($U_{mod} = 80\ \mu V$, $U = 10$ mV, $I = 25$ pA). The corresponding solid lines represent the fits using the same model as Fig. 2(b). (c) Magnetic field dependence of the normalized peak asymmetry extracted from the fits of Figs. 3(a) and 3(b) (black dots with error bar) in comparison with different models as indicated. A small offset (~ 10 mT) in the field of experimental data (black dots) is discussed in the text in detail. The inset replots the data with a focus on the low field region with a correction of the 10 mT field offset. A small overshoot can be observed at around 80 mT in the experimental data (black dots) as well as the full simulation (red dots). (d) Energy diagram of the four lowest states of the exchange coupled SOMO spin and Dy moment in magnetic field. For D_{4d} symmetry, the states of opposite magnetization become degenerate at vanishing external magnetic field [two branches for ferromagnetic (dark blue) and antiferromagnetic (light blue) alignment]. For C_4 symmetry, the degeneracy is lifted at vanishing fields due to an avoided level crossing (dashed lines).

electron undergoes switching at low fields but also the Dy magnetic moment. It, however, remains unclear, how the thermal transitions between the two Dy states happen in the presence of the large anisotropy barrier, which is much larger than the temperature [24].

Our experimental results seem—at first glance—to contradict the magnetization curves of $DyPc_2$ crystals of different Dy isotopes [13], where an essentially stable Dy magnetization was observed except for discrete fields allowing magnetization tunneling. In our case, the effect of adsorption of the molecule on the Au(111) surface comes into play. It breaks the D_{4d} symmetry, as the two Pc ligands become inequivalent, reducing the symmetry to C_4. In C_4 symmetry, additionally the Stevens operators O_4^4 and O_6^4 are allowed. As a consequence, the eigenstates become a mixture of J_z states undercutting the energy barrier. Generally, the $|+13/2\rangle$ and the $|-13/2\rangle$ states of the Dy ion are not mixed by these operators as they are not directly connected ($13 = 3 \times 4 + 1$) and the states form a Kramers

doublet. Thus, we expect a level crossing of the two states at an effective field of 0 T as depicted as solid lines in Fig. 3(d). However, when diagonalizing the complete Hamiltonian, the total angular momentum of the coupled object (Dy and unpaired electron of the SOMO) is an integer, Kramers rule fails and they start to mix by the additional Stevens operators. In this situation, the pairs of states related by time reversal symmetry show an avoided level crossing [see Fig. 3(d), dashed lines]. In the experiment this would manifest in a vanishing asymmetry near zero field in agreement with observation.

A simulation of the dI/dU spectra based on the full Hamiltonian of the zero field splitting of Dy, the exchange to the SOMO and the electron bath [30] with a realistic (electronic) temperature of 100 mK [17], was calculated and fitted to the data. The extracted normalized peak asymmetry is plotted as red dots in Fig. 3(c). Here, $B_6^4 = 0.25\ \mu$eV and $B_4^4 = 0$ were chosen. The simulation of this scenario reproduces the experimental observation even

better. Even the tiny overshoot at around ± 80 mT was reproduced qualitatively [see the inset of Fig. 3(c)]. This overshoot is due to the crossing of two other states [see Fig. 3(d)]. Note that similar results can be obtained with $B_6^4 = 0$ and $B_4^4 = 13\ \mu$eV, as both operators mix the involved states in a similar manner. Because of this ambiguity we refrain from a quantitative fit of the magnetization curve. It remains to be studied what role the nuclear spin and the hyperfine splitting play in this scenario. If the hyperfine splitting is larger than the tunneling splitting caused by the Stevens operators, a qualitative change of the behavior to that of a stable system is expected.

To conclude the discussion, the experimental results and simulations strongly suggest that the Dy magnetic moment J is not stable near vanishing magnetic fields due to the coupling with the unpaired electron and the reduced symmetry on the surface. The instability is most likely caused by an avoided level crossing. The alternative scenario of a fluctuating Dy moment can, in principle, be identified by a splitting of the two Kondo peaks into four peaks, i.e., each of the two Kondo peaks split by the external magnetic field is again split by the two possible exchange fields of Dy. Our energy resolution is, however, not sufficient to exclude this scenario. The approach to use spin-polarized STM to determine the Dy moment opens up the pathway to study clusters and artificial nanostructures of coupled DyPc$_2$ molecules realizing more complex Hamiltonians for quantum simulation. Moreover, the results provide design rules for qubits based on RE-SMMs on surfaces. The coupled electronic system under fourfold symmetry with J_z and $2 \times J_z = n \times 4 + 1$ is no longer a system with a stable $4f$ molecular spin. Instead, we suggest using coupled systems with $2 \times J_z = n \times 4 + 2$. In this case, the ground state doublet of the coupled system stays as a doublet in the presence of a fourfold anisotropy and a barrier for reversal persists even when an unpaired electron is coupled to the system. Finally, in case the nuclear spin can be utilized to stabilize the Dy moments, complex Hamiltonians of many coupled qubits can be investigated.

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[1] R. Vincent, S. Klyatskaya, M. Ruben, W. Wernsdorfer, and F. Balestro, Nature (London) 488, 357 (2012).

[2] M. Ganzhorn, S. Klyatskaya, M. Ruben, and W. Wernsdorfer, Nat. Nanotechnol. 8, 165 (2013).

[3] C. A. Goodwin, F. Ortu, D. Reta, N. F. Chilton, and D. P. Mills, Nature (London) 548, 439 (2017).

[4] C. Godfrin, A. Ferhat, R. Ballou, S. Klyatskaya, M. Ruben, W. Wernsdorfer, and F. Balestro, Phys. Rev. Lett. 119, 187702 (2017).

[5] M. Shiddiq, D. Komijani, Y. Duan, A. Gaita-Ariño, E. Coronado, and S. Hill, Nature (London) 531, 348 (2016).

[6] A. Lodi Rizzini, C. Krull, T. Balashov, J. J. Kavich, A. Mugarza, P. S. Miedema, P. K. Thakur, V. Sessi, S. Klyatskaya, M. Ruben, S. Stepanow, and P. Gambardella, Phys. Rev. Lett. 107, 177205 (2011).

[7] S. Thiele, F. Balestro, R. Ballou, S. Klyatskaya, M. Ruben, and W. Wernsdorfer, Science 344, 1135 (2014).

[8] S. Thiele, R. Vincent, M. Holzmann, S. Klyatskaya, M. Ruben, F. Balestro, and W. Wernsdorfer, Phys. Rev. Lett. 111, 037203 (2013).

[9] D. N. Woodruff, R. E. P. Winpenny, and R. A. Layfield, Chem. Rev. 113, 5110 (2013).

[10] F.-S. Guo, B. M. Day, Y.-C. Chen, M.-L. Tong, A. Mansikkamäki, and R. A. Layfield, Science 362, 1400 (2018).

[11] S. Müllegger, S. Tebi, A. K. Das, W. Schöfberger, F. Faschinger, and R. Koch, Phys. Rev. Lett. 113, 133001 (2014).

[12] C. Godfrin, S. Thiele, A. Ferhat, S. Klyatskaya, M. Ruben, W. Wernsdorfer, and F. Balestro, ACS Nano 11, 3984 (2017).

[13] E. Moreno-Pineda, M. Damjanović, O. Fuhr, W. Wernsdorfer, and M. Ruben, Angew. Chem., Int. Ed. Engl. 56, 9915 (2017).

[14] T. Komeda, H. Isshiki, J. Liu, Y.-F. Zhang, N. Lorente, K. Katoh, B. K. Breedlove, and M. Yamashita, Nat. Commun. 2, 217 (2011).

[15] T. Komeda, K. Katoh, and M. Yamashita, Prog. Surf. Sci. 89, 127 (2014).

[16] B. Warner, F. El Hallak, N. Atodiresei, P. Seibt, H. Prüser, V. Caciuc, M. Waters, A. J. Fisher, S. Blügel, J. van Slageren, and C. F. Hirjibehedin, Nat. Commun. 7, 12785 (2016).

[17] T. Balashov, M. Meyer, and W. Wulfhekel, Rev. Sci. Instrum. 89, 113707 (2018).

[18] A. Kubetzka, M. Bode, O. Pietzsch, and R. Wiesendanger, Phys. Rev. Lett. 88, 057201 (2002).

[19] O. Pietzsch, A. Kubetzka, M. Bode, and R. Wiesendanger, Phys. Rev. Lett. 92, 057202 (2004).

[20] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevLett.127.123201 for further details on the localization of the Kondo resonance, the thermal model calculation, and the synthesis of $[\text{DyPc}_2]^0$.

[21] K. Kasuga, M. Tsutsui, R. C. Petterson, K. Tatsumi, N. Van Opdenbosch, G. Pepe, and E. F. Meyer, J. Am. Chem. Soc. 102, 4835 (1980).

[22] M. Moussavi, A. De Cian, J. Fischer, and R. Weiss, Inorg. Chem. 27, 1287 (1988).

[23] N. Koike, H. Uekusa, Y. Ohashi, C. Harnoode, F. Kitamura, T. Ohsaka, and K. Tokuda, Inorg. Chem. 35, 5798 (1996).

[24] N. Ishikawa, M. Sugita, T. Okubo, N. Tanaka, T. Iino, and Y. Kaizu, Inorg. Chem. 42, 2440 (2003).

[25] N. Ishikawa, M. Sugita, T. Ishikawa, S.-y. Koshihara, and Y. Kaizu, J. Am. Chem. Soc. 125, 8694 (2003).

[26] K. Stevens, Proc. Phys. Soc. London Sect. A **65**, 209 (1952).

[27] T. Miyamachi, T. Schuh, T. Märkl, C. Bresch, T. Balashov, A. Stöhr, C. Karlewski, S. André, M. Marthaler, M. Hoffmann, M. Geilhufe, S. Ostanin, W. Hergert, I. Mertig, G. Schön, A. Ernst, and W. Wulfhekel, Nature (London) **503**, 242 (2013).

[28] T. Balashov, C. Karlewski, T. Märkl, G. Schön, and W. Wulfhekel, Phys. Rev. B **97**, 024412 (2018).

[29] K. von Bergmann, M. Ternes, S. Loth, C. P. Lutz, and A. J. Heinrich, Phys. Rev. Lett. **114**, 076601 (2015).

[30] M. Ternes, New J. Phys. **17**, 063016 (2015).

[31] P. Forrester, T. Bilgeri, F. Patthey, H. Brune, and F. Natterer, Rev. Sci. Instrum. **89**, 123706 (2018).