Atomic scale engineering of magnetic fields is a key ingredient for miniaturizing quantum devices and precision control of quantum systems. This requires a unique combination of magnetic stability and spin-manipulation capabilities. Surface-supported single atom magnets offer such possibilities, where long temporal and thermal stability of the magnetic states can be achieved by maximizing the magnetic anisotropy energy (MAE) and by minimizing quantum tunnelling of the magnetization. Here, we show that dysprosium (Dy) atoms on magnesium oxide (MgO) have a giant MAE of 250 meV, currently the highest among all surface spins. Using a variety of scanning tunnelling microscopy (STM) techniques including single atom electron spin resonance (ESR), we confirm no spontaneous spin-switching in Dy over days at ≈ 1 K under low and even vanishing magnetic field. We utilize these robust Dy single atom magnets to engineer magnetic nanostructures, demonstrating unique control of magnetic fields with atomic scale tunability.
Single lanthanide atoms adsorbed on surfaces or incorporated in molecular complexes are being pursued in recent years for applications in quantum information technology and high-density magnetic data storage. Major advances in these domains include all-electrical read-out and coherent control of their nuclear spins, the use of atomic clock transitions to protect their spins against dipolar decoherence, the discovery of single atom magnets, and the ability to manipulate their spin states.

Recently, a distinct class of molecular magnets containing a single Dy atom has even pushed the limit of operational temperatures up to 80 K by enhancing the MAE. Higher uniaxial MAE acts as a barrier against spontaneous reversal of magnetic spins, thus enhancing their magnetic lifetimes. The charge balance in such molecular magnets, however, necessitates the use of anionic counterparts, posing limitations on isolating and supporting them on a solid-state substrate. In contrast, single lanthanide atoms can be directly adsorbed on insulating MgO substrates, and due to the linear bond formed at the oxygen site, large MAE values are also expected. Among the late lanthanides, terbium (Tb), Dy, and holmium (Ho) atoms exhibit large MAE in a uniaxial crystal field (CF)1,2,3, which led to the first few lanthanide-based single-chain magnets with slow magnetic relaxation4,5. In particular, Dy ions and diatomic units deposited on MgO are predicted to possess significantly larger MAE compared to their counterparts containing Ho12. Additionally, the seminal single-atom magnet, Ho on MgO4, despite its long magnetic lifetime, lacks resilience against slow sweep rates of tip-magnetic fields16.

In this work, we report the first single-atom magnet on a surface that maintains stability at zero external magnetic field as well as against slow magnetic field sweeps, and hence can be used to create atomically localized magnetic fields. We observe magnetic stability of several days in Dy atoms adsorbed on MgO at =1 K with a giant MAE of 250 meV, using a low-temperature STM combined with single-atom ESR capability17. Using a spin-polarized tip (SP-tip) we measure random telegraph signal of the magnetic dipolar tip–magnetic stability of several Dy atoms adsorbed on MgO/Ag(001) (see Methods and Supplementary section 2). For the following, it is important to note that the SP-tips used in this work for measuring tip-field sweep ESR showed magnetic bistability in low fields ($B_{\text{ext}} < 60 \, \text{mT}$), with lifetimes typically shorter than the timescales of our measurements (see Supplementary section 2). This results in two different orientations of the tip-fields with respect to all static magnetic fields ($B_{\text{static}}$). Thus, considering only the out-of-plane projections from both $B_{\text{tip}}$ and $B_{\text{static}} = B_{\text{Dy}} + B_{\text{ext}}$, the generic condition for driving tip-field sweep ESR in Fe atoms in strong tip-field regimes ($|B_{\text{tip}}| > |B_{\text{static}}|$) can be written as (see “Methods”):

$$\frac{hf_0}{2\mu_{\text{Fe}}} = |B_{\text{tip}} + B_{\text{static}}| = |B_{\text{tip}}| + |B_{\text{Dy}} + B_{\text{ext}}|$$

Here, $h$ is Planck’s constant and $\mu_{\text{Fe}}$ is the magnetic moment of the sensor Fe atom. In the absence of any static magnetic field ($B_{\text{Dy}} = 0$ and $B_{\text{ext}} = 0$), spin resonance occurs when the tip-field induced Zeeman splitting between the two lowest lying states of the Fe atom (2$\mu_{\text{Fe}}B_{\text{tip}}$)17 matches $h\omega_0 = h\omega_0$, leading to a single resonance (Fig. 2b). However, in the presence of a static dipolar field from Dy, ($B_{\text{Dy}} > 0$ and $B_{\text{ext}} = 0$), the resonance results from two different relative alignments of $B_{\text{tip}}$ and $B_{\text{Dy}}$. Consequently, the resonance condition is satisfied at two distinct tip-fields centred around $h\omega_0/B_{\text{Dy}}$, i.e., $B_{\text{tip}} = h\omega_0/B_{\text{Dy}} - B_{\text{Dy}}$ and $B_{\text{tip}} = h\omega_0/B_{\text{Dy}} + B_{\text{Dy}}$.
Following these transitions, a series of de-excitation processes (grey arrows) cause a complete spin reversal. Data point account for standard deviation and error propagation for from multiplet calculations at \( B_{\text{ext}} = 1.5 \text{nA} \), \( V_{\text{dc}} = 100 \text{mV}, I_{\text{tip}} = 20 \text{pA} \). Spin-polarized detection of time-dependent change in apparent height \( \Delta z \) atop Dy atom exhibiting its two possible magnetic orientations \( \langle B_{\text{ext}} \rangle = 1.8 \text{K} \) for isolated Fe atoms as shown in Fig. 3a (left panel). This illustrates that the effect of the Dy dipolar field on a neighbouring Fe atom (Fig. 2b, top) is the same as having a static external magnetic field only. The positions of the two ESR peaks evolve linearly with \( B_{\text{ext}} \) for all cases (Fig. 3a). However, for the Fe–Dy pairs the two ESR peaks merge when the external magnetic field compensates the dipolar field from the Dy atom \( B_{\text{Dy}} \), thereby shifting them with respect to the isolated Fe case (Fig. 3b). From these measurements we infer the dipolar field of the Dy atom for several Fe–Dy pairs of varying interatomic distance (Fig. 3c). Note that we observe an excellent agreement between frequency sweep and tip-field sweep ESR at two different set frequencies which suggests that tip-fields are not influenced by the presence of the Dy atom (see Supplementary section 2). We fit the \( |B_{\text{Dy}}| \) values obtained from all ESR measurements to the explicit distance dependence \( B_{\text{Dy}} = \frac{\nu_{\text{Dy}}}{4\pi} \times d^{-3} \) and obtain \( \mu_{\text{Dy}} = 10.1 \pm 0.3 \mu_{\text{B}} \), in agreement with the multiplet analysis and the atomic value for Dy in gas phase.

Note that we never observe any spin-switching in Dy over days against repeated cycles of \( B_{\text{ext}} \) ramps within \( \pm 30 \text{mT} \) (see Supplementary section 3), despite an isotopic composition which bears the possibility of spin–flip transitions at low magnetic fields via hyperfine interaction in \( 161\text{Dy} \) (19% natural abundance) and \( 163\text{Dy} \) (25% natural abundance). This is in contrast to the case of single Ho atoms on MgO, where spin-switching was observed at slow magnetic field sweeps\(^{16}\). Moreover, the magnetic state of the Dy atom is also stable against high magnetic fields of 5 T and heating of at least up to 15 K (Fig. S7). Altogether, these results lead us to conclude that Dy is a single atom magnet atop O-site of MgO, even in the limit of vanishing magnetic field. The observed magnetic stability is supported by our multiplet analysis which indicates that the \( g \)-factor is essentially uniaxial with negligible transverse component (see Supplementary section 8).

Finally, we demonstrate the versatility of Dy single-atom magnets by combining their long-term magnetic stability with the ability to control their spin state by high-energy tunnelling electrons. To illustrate this, we measure tip-field sweep ESR at \( B_{\text{ext}} = 0 \text{T} \) on the Fe sensor with increasing number of surrounding Dy atoms (N), as presented in Fig. 4a. These Fe–Dy\(_N\) structures are
**Fig. 2 Zero-field magnetic stability of Dy atom.** a Schematic representation of our experimental setup for tip-field sweep ESR acquired on an Fe sensor atom at a fixed radio frequency $f_0$. Arrows with opposite orientations at the tip-apex represent the bistable nature of the out-of-plane component of the tip-field at vanishing external magnetic fields. Arrows on Fe and Dy atoms indicate respective out-of-plane magnetic moments. b Tip-field sweep ESR measured on an isolated Fe atom (green, bottom) and Fe atom in a Fe–Dy pair (blue, top) at $B_{\text{ext}} = 0 \text{T}$. Solid lines are fits to the data using a Fano–Lorentzian function (see “Methods”). Tip-field sweeps are achieved by varying the tunnelling current and thereby the tip-sample distance at a fixed dc bias ($T = 0.4 \text{K}$, $V_{\text{dc}} = -50 \text{mV}$, $V_{\text{RF}} = 27.5 \text{mV}$, $f_0 = 16.38 \text{GHz}$). The schematics highlight two opposite spin orientations of the paramagnetic tip. For a given spin-up configuration of the Dy atom, one of these tip-states satisfies the resonance condition at a lower tip–sample distance compared to the other. From the separation of the two resonance peaks we extract $B_{\text{Dy}} = 13.0 \pm 0.3 \text{mT}$. Insets show STM images of the single Fe atom and the Fe–Dy pair, where the cross indicates the position of the tip during the ESR scan and $d$ defines the distance between the Fe and Dy atoms (Image size: 2.5 × 2.5 nm², $T = 0.4 \text{K}$, $V_{\text{dc}} = 100 \text{mV}$, $I_t = 20 \text{pA}$).

**Fig. 3 Measuring the stable dipolar magnetic field of a single Dy atom.** a $B_{\text{ext}}$ dependence of tip-field sweep ESR for an isolated Fe atom (left panel), and two Fe–Dy pairs of varying distances $d$ (in middle and right panel; $T = 0.5 \text{K}$, $f_0 = 13.5 \text{GHz}$, $V_{\text{RF}} = 27.5 \text{mV}$, $V_{\text{dc}} = -50 \text{mV}$). For the case of Fe–Dy pairs, the resonance condition is satisfied at two distinct tip-fields given by, $B_{\text{tip}}^1 = \frac{\mu_0}{4\pi} \left( \frac{\mu_{\text{Fe}}}{V_{\text{RF}}} (B_{\text{ext}} + B_{\text{Dy}}) \right)$ and $B_{\text{tip}}^2 = \frac{\mu_0}{4\pi} \left( \frac{\mu_{\text{Fe}}}{V_{\text{RF}}} (B_{\text{ext}} + B_{\text{Dy}}) \right)$. Solid lines are fits to the data (see “Methods”). The colour-coded cross marks on the corresponding STM topographies indicate the position of the sensor Fe atom where the ESR measurements were taken (Image size: 3 × 3 nm², $V_{\text{dc}} = 100 \text{mV}$, $I_t = 20 \text{pA}$, $T = 0.5 \text{K}$). b ESR peak positions as a function of tip-field as extracted from the fits for all three datasets shown in (a). Error bars indicate standard deviation from fitting with equation (3). The intersection of the straight line fits provide $B_{\text{Dy}} = -5.4 \pm 0.1 \text{mT}$ (middle) and $10.4 \pm 0.4 \text{mT}$ (right), where the sign indicates two opposite Dy magnetic orientations, as in the schematics. c The dipolar magnetic field of single Dy atom $B_{\text{Dy}}$ as a function of $d$ in different Fe–Dy pairs from both tip-field sweep at two different set frequencies (red and blue) and frequency sweep (black) ESR. For tip-field sweep data, error bars indicate propagated total errors from linear fits shown in (b). For frequency sweep data, error bars indicate propagated total errors from fitting frequency sweep ESR spectra such as in Fig. S5. The solid line is a fit to the data following magnetic dipole approximation, $B_{\text{Dy}} = \frac{\mu_{\text{Dy}}}{4\pi} \times d^{-3}$, resulting in $\mu_{\text{Dy}} = 10.1 \pm 0.3 \mu_\text{B}$. 
Fig. 4 Atomic engineering of local magnetic fields. a With increasing number of surrounding Dy atoms, tip-field sweep ESR at $B_{ext} = 0$ T demonstrates the increase of $B_{Dy}$ experienced by the sensor Fe atom. Dy atoms are sequentially placed 1.15 nm apart from the central Fe atom where ESR spectra are recorded. Solid lines are fits to the data (see “Methods”). (Fe–Dy$_{N=0,1,3,4}$: $T \approx 1.3$ K, $f_0 = 21.4$ GHz, $V_{dc} = 15$ mV, $V_{RF} = -50$ mV; Fe–Dy$_4$: $T = 0.8$ K, $f_0 = 16.25$ GHz, $V_{dc} = 10$ mV, $V_{RF} = -50$ mV). Insets show corresponding STM images. (Scale bar: 1 nm; Fe–Dy$_{N=0,1,2,4}$: $V_{dc} = -100$ mV, $l_t = 20$ pA, $T \approx 1.3$ K; Fe–Dy$_4$: $V_{dc} = 80$ mV, $l_t = 20$ pA, $T \approx 1.3$ K). b Modifying $B_{Dy}$ by selectively manipulating the magnetic states of individual Dy atoms in Fe–Dy$_2$ ($T \approx 1.3$ K, $f_0 = 21.4$ GHz, $V_{dc} = 15$ mV, $V_{RF} = -50$ mV). Insets show Fe–Dy$_2$ spin schematics (red spheres: Dy spin down configuration; cyan spheres: Fe spin). For both a) and b), $B_0$ corresponds to the tip-field required for driving ESR in a reference Fe atom. Solid lines are fits to the data (see “Methods”). c, d. The dipolar magnetic field of Dy measured in different Fe–Dy$_N$ structures shown in a) and b), respectively. Additional data from an Fe–Dy$_3$ is shown in d). The solid lines in c) and d) indicate calculated values using $B_{Dy} = \frac{2\mu_0}{21.4 \text{GHz}} = \frac{2 \mu_0}{Dy}$, $d^3$. Error bars in c) and d) indicate propagated total errors from fitting ESR spectra shown in a) and b), respectively.

Methods

Sample preparation. The Ag(001) surface was prepared by several cycles of sputtering and annealing. For subsequent MgO growth, the crystal was heated to 700 K and exposed to Mg from a crucible evaporator in an oxygen partial pressure of $1 \times 10^{-10}$ mbar. Under these conditions, 40 min exposure yields an average of two monolayers of MgO. Next, the sample was cooled down to room temperature within 15 min, and was transferred to the cold STM (4 K). Prior to single atom deposition, the sample manipulator was pre-cooled by touching the sample for 20 min. Subsequently, the sample was quickly taken into an exchange chamber, where depositions of Fe and Dy were performed within a few seconds on to the cold sample using an electron beam evaporator and at a base pressure of $<7 \times 10^{-10}$ mbar. For STM measurements we used a PtIr tip, with a tip apex presumably silver-coated due to repeated indentation into the silver substrate for tip preparation. Tunnelling bias voltages were applied to the tip, however, the $V_{dc}$ values were expressed with an additional negative sign, i.e., with respect to the sample, as it is conventionally done.

ESR measurements. For the ESR measurements, an RF generator (Keysight E8257D) was used to generate the radio frequency signal, which was subsequently added to the DC bias voltage using a bias-tee (SigTek SR15D2) located outside of the vacuum chamber. A lock-in amplifier (Stanford Research Systems SR860) operating with on-off modulation at 95 Hz was used for signal detection. For conducting the tip-field sweep ESR, the RF-generator was fixed at a constant frequency and output power. Next, the STM feedback loop was kept engaged with low feedback gain and the tunnel current set point was swept. Consequently, the tip–sample distance was swept while the lock-in signal was monitored.

Given its stable magnetic orientation, the Dy atom induces a constant dipolar magnetic field on the Fe sensor. This results in two resonance peaks for all values of $B_{ext}$, which was subsequently added to the DC bias voltage using a bias-tee (SigTek SR15D2) located outside of the vacuum chamber. A lock-in amplifier (Stanford Research Systems SR860) operating with on-off modulation at 95 Hz was used for signal detection. For conducting the tip-field sweep ESR, the RF-generator was fixed at a constant frequency and output power. Next, the STM feedback loop was kept engaged with low feedback gain and the tunnel current set point was swept. Consequently, the tip–sample distance was swept while the lock-in signal was monitored.

Fitting ESR peaks. In order to determine the position of the resonance during the tip-field sweeps, we fit the experimental data shown in Fig. 2b, 3a, and 4a, b to a...
Fano–Lorentzian of the form

$$\Delta I = I_{\text{peak}} \times \frac{1}{q^2 + 1} \cdot \frac{(1 + \delta \times q)^2}{1 + \delta^2}$$  \hspace{1cm} (3)$$

Here, \( I_{\text{peak}} \) is the amplitude at the resonant tunnelling current and \( q \) is the Fano factor, arising from an additional homodyne detection of ESR\(^2\). In addition, \( \delta = \frac{B}{B_0} \), where \( B \) is the tip magnetic field for which the resonance occurs in an isolated Fe atom. The ESR signal was always normalized to the set point tunnel current for comparison. Finally for all datasets a polynomial background of degree 2 was subtracted, since the signal intensity increases with tunnel current, due to an increase of the number of read-out tunnel current electrons.

Data availability

The datasets generated during and/or analysed during the current study are available from A.S. (a.singha@fkf.mpg.de) upon reasonable request.

Code availability

There is no mathematical algorithm or custom code that is deemed central to the conclusion of this manuscript. However, the custom codes that were simply used for data analysis are available from A.S. (a.singha@fkf.mpg.de) upon reasonable request.

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Author contributions

A.S. conceived the idea. A.S., P.W., T.B., and X.Z. performed the experiments. A.S., P.W., and T.B. analysed the data. F.D. performed the multiplet analysis. T.C., F.D., H.B., and A. J.H. supervised the project. A.S. wrote the manuscript with contributions from all authors. All authors discussed the results.

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