Spintronic magnetic anisotropy
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Superparamagnetism of magnetic adatoms and molecules—preferential alignment of their spins along an easy axis—is a useful effect for nanoscale applications as it prevents undesired spin reversal. The underlying magnetic anisotropy barrier—a quadrupolar energy splitting—originates from spin–orbit interaction and can nowadays be probed by electronic transport measurements. Here we predict that in a much broader class of systems, quantum dots with spins larger than 1/2, superparamagnetism can arise without spin–orbit interaction: by attaching them to ferromagnets, a quadrupolar spintronic exchange field is generated locally. It is observable by means of conductance measurements and leads to enhanced spin filtering even in a state with zero average spin. Analogously to the spintronic dipolar exchange field, giving rise to a local spin torque, the effect is susceptible to electric control and increases with tunnel coupling as well as with spin polarization.

The growing interest in nanomagnets, for example, magnetic adatoms1 and single-molecule magnets2, is fuelled by prospects of their application in new spintronic devices whose functionality derives from their unique magnetic features3. A key property of such systems is their strong magnetic anisotropy leading to magnetic bistability, required for building blocks for nanoscale memory cells4,5, and non-trivial quantum dynamics, useful for quantum information processing6,7. In either case, operational stability of such devices hinges heavily on the height of the energy barrier opposing the spin reversal. Although recently progress in the control over the magnetic anisotropy by synthesis8, mechanical straining9, atomic manipulation10 or electrical gating11 has been made, achieving a high spin-reversal barrier still remains a challenge. Incorporating a nanomagnet into an electronic circuit may significantly alter its magnetic properties12–14, but may also be advantageous. One possible, spintronic route for manipulation of nanomagnets entails ferromagnetic electrodes and uses the spin torque due to spin–polarized scattering15 or Coulomb interaction16, magnetic analogues of the proximity effect in superconducting junctions. Here, we present another route that combines spintronics with molecular magnetism: high-spin quantum dots can acquire a significant magnetic anisotropy that is purely of spintronic origin, instead of deriving from the spin–orbit interaction, as the tunnelling to ferromagnets induces a local, quadrupolar exchange field. Besides providing an alternative approach to electrical manipulation and engineering of superparamagnetic nanomagnets, this new quantity is of key importance for the analysis of experiments that probe atoms or molecules using highly spin-polarized electrodes.

Dipolar versus quadrupolar exchange field
The origin of superparamagnetism, usually dominating the magnetic behaviour of a nanomagnet, is a magnetic anisotropy energy barrier. For instance, an adatom with a spin-degenerate ground multiplet (quenched orbital moment) is described by the generic spin Hamiltonian

$$H_{\text{eff}} = B S_z + D Q_{zz}$$

where $S_z$ denotes the component of the total spin ($S \geq 1$) along the $z$ axis, and $Q_{zz} = S_z - (1/3)S(S+1)$ is a diagonal component of the spin-quadrupole moment tensor; $B$ and $D$ are dipolar and quadrupolar fields, respectively. All parameters are in units of energy ($\hbar = |e| = 1$). If $D < 0$, the quadrupolar term prefers the axial spin states over the planar ones; that is, the spin is aligned with the $z$ axis but without favouring a particular orientation along it. For spin $S = 1$ the corresponding energy splitting is sketched in Fig. 1a. At temperatures $T < |D|$, it prevents transitions between the axial spin states through an intermediate planar state (that is, spin reversal), while maintaining the former ones as ground states. This superparamagnetism is thus of major interest for applications in which the axial states represent an information bit, and such transitions are unwanted. On the contrary, the first term in equation (1), coupling the spin dipole to the external magnetic field $B$ (chosen along the $z$ axis), does introduce a distinction between the up and down axial states. The crucial role of the $z$ axis stems from the second term of equation (1). It emerges when taking into account virtual scattering within the ground-state multiplet (e.g. of the adatom through the high-energy excited state $|e\rangle$ at energy $\Delta$, caused by the interaction of the spin ($S$) and orbital (L) angular momentum $H_{zz} = LL - S$ (see Fig. 1a). Often, only one component of orbital angular momentum has non-zero matrix elements due to ligand-field hybridization and a uniaxial intrinsic anisotropy along the $z$ axis is imposed by the negative $D = -(3/2)L^2|g|\langle L_{zz}|e\rangle^2/\Delta$. Hence, by probing the ligand environment, the atomic electrons experience a broken spin symmetry.

In spintronics a very similar situation, depicted in Fig. 1b, arises in a completely different physical setting where spin–orbit interaction is negligible. Electrons localized in a high-spin ($S > 1/2$), spin-isotropic quantum dot probe the broken spin symmetry in attached ferromagnets by virtual charge fluctuations. These fluctuations result in a spin current, which transfers spin angular momentum from the electrode to the quantum dot, resulting in a spin torque. It can be described by replacing the externally applied magnetic field in equation (1) by an effective exchange field. Specifically, we consider the set-up outlined in Fig. 1c: a quantum dot with a triplet ground state obtained by coupling an orbital level through Heisenberg interaction ($K < 0$) to an immobile, spin-1/2 impurity, causing a triplet–singlet splitting [K]. The orbital level is additionally tunnel-coupled to the ferromagnets. None of the simplifying assumptions made on the impurity, namely its immobility and its low spin value, is crucial for what follows (see Supplementary Information). The physics can be understood by

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first ignoring the second ferromagnet. Then, the dipolar exchange field \( B \) to the leading order in the tunnelling rate \( \Gamma \) decomposes into a difference of two contributions \( B_0 \) and \( B_1 \) (ref. 17):

\[
B = B_0 - B_1 \quad \text{and} \quad B_0 = \mathcal{P} \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \frac{p \Gamma f(\omega)}{\omega - \epsilon - nU/2}
\]

(2)

with \( \mathcal{P} \) standing for the principal value integral. Here, \( f(\omega) = [e^{\omega/T} + 1]^{-1} \), and \( \epsilon \) is the dot level relative to the electrochemical potential of the ferromagnet, tunable by the gate voltage. Furthermore, \( U \) is the local Coulomb interaction in the dot, \( p \) is the spin polarization of the ferromagnet, and \( W \) denotes the half-width of the conduction band. Notably, the exchange field, plotted in Fig. 2a, depends on the gate voltage linearly around \( U \). This is entirely analogous to the uniaxial spin anisotropy parameter is negative; that is, the energy of the triplet axial spin state \( |S_z = -1\rangle \) is lowered relative to the planar spin state \( |S_z = 0\rangle \), as experimentally observed in ref. 18. This is a generic feature of interacting quantum-dot spin valves\(^{19}\) if spin-polarization effects of the ferromagnets dominate\(^{20}\). Approaching the symmetry point, deep in the Coulomb blockade, processes of higher order in \( \Gamma \) become increasingly important. These are responsible for inelastic tunnelling, as well as the Kondo effect, both being primary experimental tools in atomic-scale spin detection\(^{21-23}\) and manipulation\(^5\). The interplay of such processes with the exchange field \( B(\epsilon) \) has been analysed\(^{19,24}\) and experimentally demonstrated for \( S = 1/2 \) molecular\(^23\) and carbon-nanotube\(^{18,20}\) quantum dots.

However, for high-spin quantum dots these processes result in a markedly different situation as we now explain. For our \( S = 1 \) quantum-dot example, processes of the order \( \Gamma^2 \), apart from inessential renormalization of \( B(\epsilon) \), generate an additional spin-orbit anisotropy term of the same form as the second term of equation (1). The result for the quadrupolar exchange field \( D(\epsilon) \) (Methods) is plotted in Fig. 2a. It takes a simple, approximate form in the regime \( |\epsilon + U/2| \ll U/2 \) when we neglect the excited singlet state at energy \( |K| \) and assume a large bandwidth \( W \gg U \gg T \gg \Gamma \) (as was also done in deriving equation (2)):

\[
D(\epsilon) = -B_0(\epsilon) \frac{\partial B^*(\epsilon)}{\partial \epsilon} - B_1(\epsilon) \frac{\partial B^*(\epsilon)}{\partial \epsilon}
\]

(3)

The corresponding term in equation (1) generates a quadrupolar splitting. We find that in the Coulomb blockade regime this \( D \) parameter is negative; that is, the energy of the triplet axial spin states \( |S_z = \pm 1\rangle \) is lowered relative to the planar spin state \( |S_z = 0\rangle \), as in Fig. 1b. This is entirely analogous to the uniaxial spin anisotropy typical of magnetic adatoms or molecules (see Fig. 1a). However, this anisotropy is induced by the proximity of the ferromagnet and exhibits the characteristic properties of a spintronic exchange field: it is electrically tunable by the gate voltage as shown in Fig. 2a, and scales as \( D \propto p^2 \Gamma^2 \). It is thus enhanced with increasing tunneling coupling \( \Gamma \), similar to the Kondo effect (see below), but in contrast, it is also enhanced with increasing spin polarization \( p \), which suppresses the Kondo effect.

Importantly, Fig. 2a demonstrates that the quadrupolar field is a symmetric function of the gate voltage, \( D(\epsilon) = D(-\epsilon - U) \), and therefore does not necessarily vanish at the symmetry point (the electron and hole contributions to equation (3) add up, unlike in equation (2)). Expanding \( B(\epsilon) \) and \( D(\epsilon) \) linearly around the symmetry point,

\[
B(\epsilon) \approx -\frac{2}{\pi} \frac{p \Gamma}{U} \left( \frac{U/2}{U} \right) \ln \frac{2W}{U} = D^* \quad \text{(linear)}
\]

(4)

\[
D(\epsilon) \approx -\frac{1}{\pi^2} \left( \frac{p \Gamma^2}{U} \right) W \ln \frac{2W}{U} \quad \text{(constant)}
\]

(5)
we thus obtain an all-spintronic superparamagnet described by equation (1) with a constant anisotropy $D^*$ and a magnetic field that is linearly tunable by the gate voltage through $\epsilon$ (see Fig. 2b). Close to the symmetry point, the quadrupolar field dominates over the dipolar one in the gate voltage range $|\epsilon + U/2| \ll \delta \epsilon := p \Gamma \ln(2W/U)/2\pi$ proportional to the width $\Gamma$ of the low-temperature Coulomb peaks. Accordingly, high-spin quantum-dot spin valves exhibit a tunable interplay of spintronic and nanomagnetism that is not possible for low-spin quantum dots. This, in turn, opens the possibility of fast all-electrical operations involving the spin, which are challenging for adatoms and single-molecule magnets.

**Spectral signatures of spintronic quadrupolar splittings**

On the basis of the perturbative results (4)–(5) we expect a clear experimental signature of the spintronic quadrupolar field for strong tunnel coupling $\Gamma$. The above considerations are readily extended to the case of a junction of two ferromagnets with voltage bias $V_b$ and parallel polarizations (Methods). The spintronic fields $B$ and $D$ now also acquire a dependence on the bias voltage, which is, however, negligible in the situation under discussion (see Supplementary Information). To address the strong tunnelling regime and to better estimate the achievable magnitude of $D$ we calculate the equilibrium, spin-resolved local density of states including the singlet excited state at finite energy $|K|$ that we neglected so far, and taking into account the Kondo effect, which also gains importance with increasing $\Gamma$ at low temperature.

To set the stage, we show in Fig. 3a the result for a spin $S = 1/2$ quantum-dot spin-valve model (obtained by setting $K = 0$), successfully used to analyse the spectroscopy of the dipolar exchange field \(^{14,20}\). Unlike for non-magnetic electrodes, a Kondo peak forms only at the symmetry point where the exchange field $B$ induced by the ferromagnets vanishes \(^{19,22,26}\). The finite-temperature precursor of the Kondo peak in Fig. 3a exhibits the measured, gate-voltage dependent splitting \(^{18}\).

For a high-spin $S = 1$ ground state (that is, $K < 0$), instead of a peak, we find in Fig. 3b a pronounced gap, which linearly increases as the gate voltage is detuned from the symmetry point. This indicates a definite spin excitation, even close to the symmetry point where the influence of a dipolar exchange field $B$ on the high-spin quantum dot is negligible. As illustrated in Fig. 2b, the observed excitation as a function of $\epsilon + U/2$ is the telltale signature of uniaxial spin anisotropy of the type predicted by equations (1) and (3). By relating the gate voltage through equation (4) to the magnetic field, this signature is seen to coincide with that of intrinsic anisotropy discussed theoretically and observed experimentally in the transport through real magnetic adatoms and molecules \(^{10,11,27}\). The failure to close the gap at the symmetry point in Fig. 3b corresponds to a so-called zero-field splitting \(^{28}\) of such systems. During the excitation the spin transits from the lowest axial state $|S_z = \pm 1\rangle$ into the planar state $|S_z = 0\rangle$, see Fig. 2b. Direct transitions between the axial spin levels are spin-forbidden and do not show up in Fig. 3b. Importantly, the different energy units on the left and right $\omega$-axis in Fig. 3c,d reveal that the DM-NRG gap is indeed of spintronic origin: it scales with $\Gamma$ and $p$ as predicted by equations (4)–(5) for $|B| \gg |D|$ and $|B| \ll |D|$, respectively. We note that only for much stronger coupling the Kondo effect in Fig. 3b reinstates the characteristics similar to that in Fig. 3a. Finally, the quadrupolar gap can also be extracted from the temperature dependence of the transport quantities (see Supplementary Fig. S6).
Finally, the spintronic quadrupolar exchange field can also be used to enhance the linear-response spin filtering of electrons transported through a quantum dot with zero dipolar exchange field and average spin. Figure 4a presents the linear conductance as a function of the tunnel coupling $I'$ for fixed, intermediate temperature at the symmetry point where $\langle S \rangle = 0$ (see Supplementary Information). We observe that with increasing spin polarization $\rho$ of the ferromagnets, the value of $I'$ for which the Kondo unitary conductance is reached strongly increases. This shift is caused by the spintronic anisotropy, evidenced by the finite quadrupolarization $\langle Q_{zz} \rangle > 0$ in Fig. 4b, which arises when $|D^j(\Omega)| \gg T$—well before the Kondo effect sets in. Interestingly, the conductance spin polarization in Fig. 4c shows a corresponding peak that develops into a 100%-plateau for $p$ close to, yet still less than, 1. However, even for 50% polarized ferromagnets ($p = 0.5$), the conductance spin polarization can be almost doubled if the temperature is further reduced (see Supplementary Fig. S10).

This amplification of the conductance spin polarization is a hallmark of the exchange quadrupolar field: in a broad, intermediate regime of $I'$-values the spintronic anisotropy gap $|D^j| \gg T$ makes the planar spin state $\langle S_z \rangle = 0$ inaccessible at low energy, as depicted in Fig. 4(ii). In consequence, tunnelling-induced spin-flip processes between the axial states $\langle S_z \rangle = \pm 1$ (Kondo effect) are strongly suppressed, greatly enhancing the conductance spin polarization (see Supplementary Information). Such a behaviour is absent in the results for $S = 1/2$ shown for comparison as thin curves in Fig. 4c: in this case, spin reversal does not involve a planar state.

This enhanced spin-filtering effect illustrates a promising synergy of spintronics and nanomagnetism. The spintronic anisotropy may offer new possibilities for combining tools and insights from these two research fields. As we show in the Supplementary Information, one could think of using the quadrupolar field as a new means for fast, electronic control of nanomagnetic memory cells, allowing even for on-demand bistability. Furthermore, in contrast to intrinsic anisotropy, the spintronic anisotropy can also be turned off magnetically by switching the spin valve to the antiparallel configuration. Finally, it is interesting to consider whether spintronic anisotropy may be understood in a broader context: just as the dipolar exchange field term in equation (1), expressing the spin torque, can be seen as a part of the dipole current, so, perhaps, the quadrupolar exchange field term may relate to a spin-quadrupole current.

**Methods**

The model of a high-spin quantum-dot spin valve in Fig. 1c consists of a single spin-degenerate orbital level, tunnel-coupled to two ferromagnets, and involving the local isotropic Heisenberg exchange coupling $K < 0$ to an immobile $S = 1/2$ impurity. Electron tunnelling through the junctions is assumed to be symmetric and spin conserving.

**Real-time diagrammatic perturbation theory.** By integrating out the ferromagnets held at different equilibria we obtain a stationary quantum kinetic equation for the reduced density operator $\hat{\rho}(t) = 0 = -i[H_{\text{dot}}, \hat{\rho}] - i\Sigma \hat{\rho}$, where $\Sigma$ is the zero-frequency kernel expanded formally in powers of $I'$. We rewrite $\Sigma = \Sigma_{\text{core}} + \Sigma_\rho$, where $\Sigma_{\text{core}}$ is the renormalization of the Hamiltonian $H_{\text{dot}}$ including both orders $I'$ and $I''$, which can be identified diagrammatically and then calculated. See Supplementary Section SIIA for a more detailed account. In the Coulomb blockade regime, $H_{\text{dot}}$ reduces to a trivial constant and $\Sigma_{\text{core}}$ is given by equation (1), where $D$ for a single electrode has the form

$$D = \frac{\Gamma'^2}{4} R e \int \frac{d\omega}{\pi} \frac{1}{\pi} \left[ \frac{1 - f(\omega_n)}{\omega_0 - \omega_0 + i\delta} \right]^2$$

(6)

This was used to plot Fig. 2a. In the limit $U \ll W$, the expression reduces to the analytic result (3), from which equation (5) follows. As mentioned in the text,
one can extend the results to the case of two electrodes \( r = L, R \), with respective electrochemical potentials \( \mu_{\alpha} = \pm V_i/2 \): replace in equation (2) \( f(\omega) = f(\omega) = \frac{1}{[e^{\frac{\omega - (\mu - V_i)/2}{k_B T}} - 1]^2 + \text{sum over } r \). Equation (3) remains valid as long as \( V_i < U \).

Finally, we recall that the dipolar exchange field in order \( T^c \) vanishes at the symmetry point \( \epsilon = -U/2 \), see equation (4) and Fig. 2a. This also holds for irrelevant higher-order corrections in \( T^c \) to \( B \), as the zero average spin, obtained from DM-NRG calculations at the symmetry point, demonstrates for any values of \( p \) and \( \Gamma^c \).

DM-NRG. We use the flexible DM-NRG (ref. 35) approach to numerically calculate the spin-resolved equilibrium spectral function \( a_{\sigma} (\omega) \) \( \sigma = \uparrow, \downarrow \), referred to also as local density of states, of the orbital level for parallel polarization of the ferromagnets. From this we compute the spin-resolved linear-response conductance

\[
G_{\sigma} = \frac{2e^2}{h} \pi \Gamma (1 + \eta_p) \int d\omega \left[ \frac{\delta f(\omega)}{\delta \omega} \right] a_{\sigma}(\omega)
\]

(7)

where \( \eta_p = \pm 1 \). Moreover, by approximating \( df/d\omega \propto A(\omega) \) at low \( T \) and low, but finite bias voltage \( V_i = \omega \) with the symmetrized, dimensionless equilibrium spectral function \( A(\omega) = \pi \Gamma \sum (1 + \eta_p) p[a_{\sigma}(\omega) + a_{\sigma}(-\omega)]/2 \), we can infer useful qualitative conclusions about the differential conductance, taking into account known limitations of this approximation. Further details can be found in Supplementary Section SII.

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Author contributions

M.R.W. conceived the idea. M.H. and M.M. performed the analytic and numerical calculations, respectively. M.H. provided M.M. and M.R.W. with fitting formulas for the physical analysis of DM-NRG results. M.M. prepared the initial manuscript. All authors contributed to writing the manuscript.

Additional information

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Competing financial interests

The authors declare no competing financial interests.