Magnetic anisotropy of the noncollinear antiferromagnet IrMn₃

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The magnetic anisotropy of antiferromagnets plays a crucial role in stabilizing the magnetization of many spintronic devices. In noncollinear antiferromagnets such as IrMn₃, the symmetry and temperature dependence of the effective anisotropy are poorly understood. Theoretical calculations and experimental measurements of the effective anisotropy constant for IrMn differ by two orders of magnitude, while the symmetry has been inferred as uniaxial in contradiction to the assumed relationship between crystallographic symmetry and temperature dependence of the anisotropy from the Callen-Callen law. In this Rapid Communication, we determine the effective anisotropy energy surface of L₁₂-IrMn₃ using an atomistic spin model and constrained Monte Carlo simulations. We find a unique cubiclike symmetry of the anisotropy not seen in ferromagnets and that metastable spin structures lower the overall energy barrier to a tenth of that estimated from simple geometrical considerations, removing the discrepancy between experiment and theory. The temperature scaling of the anisotropy energy barrier shows an exponent of 3.92, close to a uniaxial exponent of 3. Our results demonstrate the importance of noncollinear spin states on the thermal stability of antiferromagnets with consequences for the practical application of antiferromagnets in devices operating at elevated temperatures.

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I. INTRODUCTION

The magnetic anisotropy of antiferromagnetic (AF) materials plays a key role in the stability of many spintronic devices [1–5] and exchange bias effects [6–8]. Recently, interest in the properties of AF materials has increased due to their emerging applications in AF spintronic [3,5] and neuromorphic computing devices [9] where the antiferromagnet is the active element. The magnetic anisotropy energy density as determined experimentally is a free-energy difference between maximum and minimum on the free-energy surface where the temperature variation of the anisotropy arises from spin fluctuations. For clarity, we refer to the intrinsic quantity, determined from ab initio calculations, as the magnetocrystalline anisotropy energy (MAE) and the experimental temperature-dependent free-energy density as the magnetocrystalline anisotropy constant K. Magnetic anisotropy is usually classified by symmetry in expansions of spherical harmonics with azimuthal and rotational components [10] describing uniaxial and cubic forms of the anisotropy. The temperature dependence of the anisotropy is intrinsically related to the order of the harmonics [10,11] and is well understood for ferromagnets. In contrast, the magnetic anisotropy of antiferromagnets is poorly understood due to the difficulty in experimental measurements, the complexity of the materials, and noncollinear magnetic structure.

Iridium manganese (IrMn) is the material chosen for many AF spintronic devices due to its high thermal stability and large exchange bias field. In devices, the ordering and composition is tuned for optimal performance but here we focus on the L₁₂ ordered IrMn₃ phase due to the existence of extensive experimental [12,13] and theoretical [14–16] data. Theoretical calculations by Szunyogh et al. [14] found an extremely large second-order MAE for IrMn₃, leading to a predicted magnetocrystalline anisotropy energy density of the order of 3 × 10⁷ J/m³ at 0 K. Vallejo-Fernandez et al. [17,19] calculated a value of the anisotropy constant K = 6.2 × 10⁸ J/m³ at 300 K or K = 14.8 × 10⁷ J/m³ at 0 K, almost two orders of magnitude lower than the expected value from theoretical calculations [14]. The experimental estimate of the anisotropy constant of IrMn is sensitive to the value of the switching attempt frequency (f₀) in the Arrhenius Néel law given by

\[ 1/\tau = f_0 \exp \left( -\Delta E/k_BT \right) \]  

where τ is the relaxation time, \( \Delta E \) is the energy barrier, \( k_B \) is the Boltzmann constant, and T is the temperature. Originally, Vallejo-Fernandez et al. used a value of \( f_0 = 10^9 \text{s}^{-1} \) [17] but more recent estimates suggest values closer to \( f_0 = (2.1 \pm 0.4) \times 10^{12} \text{s}^{-1} \) [19]. Determination of the exact value of the attempt frequency is therefore critical to determining the effective anisotropy in AF materials.

A further unresolved problem relates to the symmetry of the IrMn₃ anisotropy. Vallejo-Fernandez [17] and Craig et al. [20] calculated the anisotropy by fitting to the temperature dependence of the magnetization using a Callen-Callen [11] power law \( K_{\text{AF}}(T)/K_{\text{AF}}(0) = (n_{\text{AF}}(T)/n_{\text{AF}}(0))^l \), with \( n_{\text{AF}} \) the AF sublattice magnetization. The exponent \( l \) reflects the symmetry of the anisotropy, which itself generally reflects that of the lattice. Agreement with experimental measurements [17] requires an exponent of \( l \sim 3 \) (uniaxial) rather than

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l ∼ 10 (cubic anisotropy). Szunyogh et al. [14] showed that the local energy surface for individual spins is uniaxial by rotating the triangular ground state about the (111) direction. Both experiment and theory agree that the anisotropy has an approximately uniaxial form. However, this contradicts the predicted relationship between crystallographic symmetry and the temperature dependence of the anisotropy from the Callen-Callen and Zener relations [10,11].

In this Rapid Communication, we resolve this apparent contradiction by determining the equilibrium anisotropic free-energy surface and by calculation of the temperature dependence and scaling of the effective anisotropy. We find that the anisotropy of IrMn3 possesses a unique symmetry neither uniaxial nor cubic in nature and has an unusual scaling exponent of \( t = 3.92 \), not seen for ferromagnetic materials.

II. METHOD

To study the anisotropy of L12-IrMn3 we use an atomistic spin model where the energy of the system is defined using the spin Hamiltonian

\[
\mathcal{H} = - \sum_{i<j} J_{ij} \boldsymbol{S}_i \cdot \boldsymbol{S}_j - \frac{k_B}{2} \sum_{i \neq j} (\boldsymbol{S}_i \cdot \mathbf{e}_{ij})^2, \tag{2}
\]

where \( \boldsymbol{S}_i \) is a unit vector describing the spin direction on Mn site \( i \), \( J_{ij} \) is the Néel anisotropy constant, and \( \mathbf{e}_{ij} \) is a unit vector from site \( i \) to site \( j \). \( z \) is the number of nearest neighbors and \( J_{ij} \) is the exchange interaction. The effective exchange interactions \( J_{ij} \) were limited to nearest \( (J_{nn}^m = -6.4 \times 10^{-23} \text{ J/link}) \) and next-nearest \( (J_{nn}^m = 5.1 \times 10^{-21} \text{ J/link}) \) neighbors [21]. In IrMn, the magnetocrystalline anisotropy arises from the large spin-orbit coupling between Mn and Ir sites [14]. Here we map the local anisotropies at each Mn site to a Néel pair anisotropy model [21,22] which gives exact agreement with the ab initio calculations [14]. The Néel model reflects the local site symmetry to give the correct easy axes for each Mn site and by performing coherent spin rotations as in Ref. [14], we find the same angular dependence of the anisotropy energy.

III. RESULTS

To verify the model we calculated the ground-state spin structure of ordered L12-IrMn3 using a Monte Carlo metropolis algorithm with the adaptive update method [23,24] and implemented in the VAMPIRE software package [25]. The 8 × 8 × 8 nm\(^3\) system was initially equilibrated at a temperature of 1500 K (above the Néel temperature) to thermalize the spins. The system was then cooled to 0 K using a linear cooling function over 10\(^6\) Monte Carlo steps to find the ground-state spin configuration. In agreement with previous experimental [12,13] and ab initio results [14], we find that ordered L12-IrMn3 has a triangular (T1) spin structure where the magnetic moments lie parallel to the [111] planes as shown in Fig. 1. There are eight [111] planes and, by symmetry, IrMn has eight magnetic ground states.

The energy barrier separating two ground states is the minimum energy path for the spins to rotate between them and defines the effective anisotropy and the thermal stability.

To calculate the energy barrier, we use the constrained Monte Carlo algorithm to determine the free-energy surface and the energy barrier to magnetic reversal [26]. Here, we constrain the direction of magnetization of a single Mn sublattice while allowing all other spins to relax to obtain the equilibrium spin structure with a constraint applied. By scanning all angles \((\theta, \phi)\), the energy surface is obtained. For each value of \( \theta \) and \( \phi \), the (8 nm)\(^3\) system was initially heated to 1500 K to thermalize the spins and then cooled to 0 K. Due to the constraint, the system cannot reach a full equilibrium and so the total internal torque \( \tau \) is nonzero and given by

\[
\tau = -\mathbf{M} \times \frac{\partial \mathcal{F}}{\partial \mathbf{M}}, \tag{3}
\]

where \( \mathcal{F} \) is the Helmholtz free energy which is a function of \( \mathbf{M} \). Since \( \mathcal{F} \) cannot be computed directly, we reconstruct it by numerical integration of the torque

\[
\mathcal{F} = \mathcal{F}_0 + \int_{M}^{M'} (\mathbf{M} \times \mathbf{T}) \cdot d\mathbf{M}', \tag{4}
\]

taken along any path between two points on the energy surface. The computed energy surface at 0 K is shown in Fig. 2(a) and has a complicated structure with four minima. The energy minima lie at \( \phi \sim \pm 56^\circ \), corresponding to the expected easy directions of the constrained sublattice. To calculate the energy barrier between two adjacent minima, we compute the minimum energy path between them as shown in Fig. 2(b). The calculated 0 K anisotropy is 1.78 × 10\(^6\) J/m\(^3\), which is an order of magnitude lower than that for rigid rotation of spins calculated by Szunyogh et al. [14] and has massively reduced the disparity between the experiment and theory with this result being less than 10% off the experimental measurement. The surprising reduction arises due to a bobbing motion of the unconstrained spins which results from the competition between the exchange and anisotropy.
energies leading to small deviations from the ground-state spin structure when the AF spins are rotated. This is particularly relevant to macroscopic approximations of AF materials with Néel vectors where the sublattices are always assumed to have a fixed local spin structure. The remaining difference in the values of the effective magnetic anisotropy could be due to different ordering or defects in the experimental samples, but our results finally resolve the large disparity between the theoretically calculated and experimentally measured magnetic anisotropy of IrMn₃. We note that, although the energy surface illustrated in Fig. 2(a) has an unusually complex form, the minima themselves exhibit a fourfold symmetry, characteristic of cubic rather than uniaxial anisotropy. The question remains how to resolve the apparent contradiction with the experimental data of Vallejo-Fernandez et al. [17] and its requirement of a magnetization scaling exponent consistent with uniaxial symmetry.

To resolve this discrepancy, we now investigate the temperature dependence of the anisotropy constant to calculate the scaling exponent. The energy surfaces and minimum energy path were calculated for temperatures between 0 K and 350 K as shown in Fig. 3(a). The absolute free energy increases with temperature due to spin fluctuations but the free-energy barrier decreases. In Fig. 3(b), we plot the power-law dependence of the effective energy barrier as a function of the magnetization and find an unusual exponent of \( l = 3.92, \pm 0.14 \). The exponent is closer to a uniaxial exponent of \( l = 3 \), matching the experimental observations but deviates from this ideal value due to the complex symmetry of the anisotropy energy surface. We also note that the specific scaling exponent is dependent on the strength of the anisotropy, and for weaker anisotropy tends toward exponent of \( l = 3 \), which may be seen in similar noncollinear magnets such as PtMn₃. We conclude that the magnetic anisotropy of L₁₂-IrMn₃ possesses a close to uniaxial temperature dependence in direct contradiction with the usual Callen-Callen power laws and cubic nature of the crystal [11].
Finally, another problem in calculating the anisotropy comes from the experimentally determined values of the anisotropy constant; we consider the basis of the experiments [17,18], where the attempt frequency $f_0$ is a critical parameter in calculating the effective anisotropy. Having determined the precise energy barrier at an elevated temperature, we are now able to compute the attempt frequency using atomistic spin dynamics. We simulate the dynamic behavior using the stochastic Landau-Lifshitz-Gilbert (sLLG) equation [24,27]

$$\frac{\partial S_i}{\partial t} = -\frac{\gamma}{1+\lambda^2}[S_i \times B_{\text{eff}} + \lambda S_i \times (S_i \times B_{\text{eff}})], \quad (5)$$

where $\lambda$ is the Gilbert damping constant and $[\gamma]$ is the gyromagnetic ratio. The effective field $B_{\text{eff}}$ is calculated as the derivative of the spin Hamiltonian with respect to the local spin moment plus a random thermal field ($B_{\text{eff}} = -\mu S_0^{-1}\partial H'/\partial S_i + B_{\text{th}}$), where $B_{\text{th}} = \Gamma(t)\sqrt{2\Delta k_B T/\gamma\mu S_0^{-1}}$ and $\Gamma$ is a 3D random Gaussian distribution. The sLLG equation is integrated using a second-order predictor corrector Heun scheme [24].

We determined the attempt frequency by calculating the transition rate just below the blocking temperature of the antiferromagnet. Due to the giant anisotropy of IrMn$_3$ and limited time accessible by simulations, we simulate a small sample (1.5 nm$^3$) which has a blocking temperature of $T_B = 101.5K$ for a timescale of 0.1 ns. To precisely calculate the energy barrier for this system at 100 K, we use the same method as above to calculate the energy surface. The time-dependent dynamics of the magnetization for a single sublattice is shown in Fig. 4.

As the temperature is just below the blocking temperature, the IrMn switches between stable states giving a time-dependent form similar to telegraph noise. Over a total simulation time of 100 ns, the total number of transitions was calculated and divided by the total simulation time. The frequency of the transitions is dependent on the magnitude of the damping constant, which is typically in the range 0.01 to 1 for materials with large spin-orbit coupling. The simulation was repeated for damping constants within this range to determine the variation in the attempt frequency, giving $f_0$ values between 0.1 and $4 \times 10^{12}$ Hz, shown in Fig. 4. The simulated values are of the same order as the experimentally determined value [19] and provide reasonable bounds for the attempt frequency for noncollinear antiferromagnets.

**IV. DISCUSSION**

Applying constrained minimization and spin dynamics simulations, we have determined the symmetry and effective temperature-dependent anisotropy and relaxation dynamics of IrMn$_3$, one of the most technologically important noncollinear AF materials. We find that the anisotropy energy surface is unusually complex and find a scaling exponent different from the expectations of Callen-Callen theory despite the presence of cubic crystal symmetry and localized uniaxial anisotropy at atomic Mn sites. Metastable spin structures are shown to lower the overall energy barrier to a tenth of that estimated from simple geometrical approximations. Spin dynamics calculations reveal an exceptionally high attempt frequency in IrMn$_3$ of between $f_0 = 0.1$ and $4 \times 10^{12}$ s$^{-1}$; a value three orders of magnitude larger than the typical value chosen for ferromagnets of $10^9$ s$^{-1}$. Considering a specific value for the damping constant of $\lambda \approx 0.1$ gives a comparable value of the attempt frequency with respect to the measured experimental value [19] of 2.4 THz. We have therefore resolved the outstanding discrepancy between theory and experiments calculating a value for the anisotropy energy within 10% of the experimental value.

From the structure of the zero Kelvin energy surface we conclude that the magnetic anisotropy of L1$_2$-IrMn$_3$ possesses a unique symmetry not seen for ferromagnets, yet consistent with the expected relationship to the crystal symmetry by rotation of the ground-state spin structure. We find that the scaling of the anisotropy is similarly unusual with an exponent of 3.92, which is closer to uniaxial magnetization scaling of the anisotropy despite the near-cubic symmetry. The resolution of this apparent contradiction (and the associated experimental measurements) is as follows. Although the anisotropy is shown to have cubiclike symmetry, the scaling arises from the spin fluctuations which reflect the local uniaxial environment of individual spins. This is an intriguing and unusual separation of the fundamental origin of the MAE and the temperature dependence of the anisotropy.
Although we have focused on IrMn$_3$ we expect that other noncollinear antiferromagnets such as MnPt and MnFe will exhibit similarly complex temperature-dependent magnetic anisotropy. This is likely to be strongly affected by composition and ordering which will disrupt the local anisotropy energy surface at different atomic sites. Our results have important consequences for applications of antiferromagnets in determining their thermal stability and dynamic properties and provide an established methodology for determining the effective magnetic anisotropy at elevated temperatures. This is particularly important for emerging applications in neuromorphic computing and AF spintronics where the long-term stability of the antiferromagnet is critical to device operation. Further investigation may yield different classes of antiferromagnets with unusual temperature-dependent properties.

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