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Coercive field of a polycrystalline ferrimagnet with uni-axial anisotropy

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Abstract

Unlike the coercive field H_c of a bulk ferrimagnet, which diverges at the compensation temperature T_{comp} , the coercive field of a polycrystalline ferrimagnet with uni-axial anisotropy is shown to have a minimum at T_{comp} . Despite this behavior, the field required for domain-wall motion still diverges at the compensation temperature. These ideas are used to treat a ferrimagnetic class of molecule-based magnets, the bimetallic oxalates, that exhibit a minimum coercivity at T_{comp} .

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Till recently, the coercive field H_c of every known bulk ferrimagnet was found to diverge at its magnetic compensation temperature T_{comp} , where the magnetic moments of the two or more sublattices cancel each other [1]. This effect has widespread technological applications and is easily explained by a simple calculation of the domain-wall energy [2], which predicts that H_c is inversely proportional to the net ferrimagnetic moment M_s . However, recent measurements of the coercive field of a polycrystalline organic ferrimagnet [3] reveal a minimum rather than a maximum at the compensation temperature. The only other known materials where the coercivity is predicted [4] and observed [5] to exhibit a minimum at T_{comp} are ferrimagnetically coupled multilayers. In this paper, we demonstrate that the coercivity of a polycrystalline ferrimagnet with uni-axial anisotropy should likewise have a minimum at T_{comp} . After sketching a very basic derivation of the coercive field for a polycrystalline ferrimagnet with uni-axial anisotropy, we evaluate the coercivity of the polycrystalline, molecule-based magnet studied in Ref. [3].

Consider a single-crystal ferrimagnet with uni-axial anisotropy along z -axis. If this ferrimagnet is fully magnetized with $\mathbf{M} = -M_s \mathbf{z}$, then the condition for domain-wall motion in a magnetic field \mathbf{H} is given by $H_z = H \cos \theta \geq H_c^{(0)}$, where $H_c^{(0)}$ is the coercive field along

z -axis and $\{\theta, \phi\}$ are the spherical angles of \mathbf{H} with respect to that axis. Due to the components of \mathbf{H} perpendicular to the ferrimagnetic moments, there will also be an induced moment $\chi_{\perp} H \sin \theta$ in the xy plane. So the total moment can be written

$$\mathbf{M}(\theta, \phi) = -M_s \mathbf{z} + 2M_s \mathbf{z} \Theta(H \cos \theta - H_c^{(0)}) + H \chi_{\perp} \sin \theta (\cos \phi \mathbf{x} + \sin \phi \mathbf{y}), \quad (1)$$

where $\Theta(x) = 1$ if $x > 0$ and 0 otherwise. After integrating over all angles with $0 \leq \theta \leq \pi/2$, we find that the average magnetization along the field direction of a collection of particles, all fully magnetized in the southern hemisphere, is

$$M = M_s \left\{ -\frac{1}{2} + \left(1 - \left(\frac{H_c^{(0)}}{H} \right)^2 \right) \Theta(H - H_c^{(0)}) \right\} + \frac{2}{3} H \chi_{\perp}. \quad (2)$$

This expression gives a remanent magnetization of $-M_s/2$ in zero field. Notice that the slope of $M(H)$ for fields $H \gg H_c^{(0)}$ is $\tilde{\chi}_{\perp} = 2\chi_{\perp}/3$. Unlike in an antiferromagnet, $\tilde{\chi}_{\perp}$ is weakly temperature dependent due to the different moments on the two or more sublattices of the ferrimagnet.

The coercive field H_c of a polycrystal is defined as the field where $M = 0$. As implied by Eq. (2), H_c depends on whether $\alpha \equiv M_s/(2H_c^{(0)}\tilde{\chi}_{\perp})$ is larger or smaller than 1. For M_s small enough that $\alpha < 1$, $h \equiv H_c/H_c^{(0)} = \alpha$. But for a larger magnetization with $\alpha > 1$, h must be solved from the

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cubic equation

$$h + \alpha(1 - 2/h^2) = 0 \quad (3)$$

which is satisfied by $h = 1$ when $\alpha = 1$. The hysteresis loops in these two regimes are plotted in Fig. 1. Notice that $h > 1$ only when $\alpha > 1$. So the coercive field is large enough to move the domain walls only when $M_s > 2H_c^{(0)}\tilde{\chi}_\perp$. In the vicinity of T_{comp} with $\alpha < 1$, the perpendicular susceptibility determines the coercive field $H_c = M_s/(2\tilde{\chi}_\perp)$. A similar mechanism is responsible for the minimum coercive field at T_{comp} in ferrimagnetic multilayers [4].

Irreversible domain-wall motion only occurs along the curved portions of the hysteresis loops in Fig. 1 with $|H| > H_c^{(0)}$. The hysteresis loops are reversible along the straight-line paths in Fig. 1, where the particles are magnetized perpendicular to their local z -axis. So with $M = -M_s/2$ in zero field, the magnetization can be reversed up to fields much larger than H_c in the vicinity of T_{comp} .

All of the moments oriented along the local z -axis of each particle will contribute to χ_\perp . But due to disorder, only a fraction x_d will contribute to the ferrimagnetic moment $M_s(T)$. So we write $M_s(T) = x_d M_0(T)$, where $M_0(T)$ is the net ferrimagnetic moment for a perfect single crystal at temperature T . The maximum coercive field below T_{comp} is given by the conditions $H_c = H_c^{(0)}$ and $\alpha = 1$ so that $H_c^{\text{max}} \approx x_d M_0/(2\tilde{\chi}_\perp)$.

To demonstrate these ideas, we now consider the class of layered molecule-based magnets that were found to exhibit a minimum coercivity at T_{comp} in Ref. [3]. Bimetallic oxalates [6] are salts with the chemical formula

$A[M(\text{II})M'(\text{III})(\text{ox})_3]$, where A is an organic cation that separates the negatively charged metallic layers. Each of the metallic layers contains two different metal atoms in an alternating honeycomb structure. Neighboring metal atoms $M(\text{II})$ with valence $+2$ and $M'(\text{III})$ with valence $+3$ are bridged by the oxalate molecule $\text{ox} = \text{C}_2\text{O}_4$ with valence -2 . Most commonly, $M(\text{II}) = \text{Mn, Ni, Fe, Co, Cu,}$ or Zn and $M'(\text{III}) = \text{Cr, Ru,}$ or Fe . Depending on the metal atoms, a single bimetallic layer can be either ferromagnetic or ferrimagnetic ($M(\text{II})$ and $M'(\text{III})$ moments parallel or anti-parallel) with magnetic moments pointing out of the plane. While it does not change the sign of the exchange coupling, the organic cation A does affect the overall behavior of the system. With the appropriate cation, bimetallic oxalates can be optically activated [7], metallic [8], or disordered [9,10].

In Ref. [3], Coronado et al. studied an $\text{Fe}(\text{II})\text{Fe}(\text{III})$ bimetallic oxalate with $A = \text{NBu}_4$. Depending on the cation, some $\text{Fe}(\text{II})\text{Fe}(\text{III})$ compounds exhibit magnetic compensation due to the cancellation of the moments on the $S = 2$ $\text{Fe}(\text{II})$ and $S' = \frac{5}{2}$ $\text{Fe}(\text{III})$ sublattices [11]. The compound studied by Coronado et al. has a transition temperature of about 45 K and a compensation temperature of about 30 K.

The degree of disorder in the $\text{Fe}(\text{II})\text{Fe}(\text{III})$ bimetallic oxalates is rather high. With $|\langle L_z \rangle| \approx 0.23$ on the $\text{Fe}(\text{II})$ sites (see below), a low-temperature moment $M_0(0) = 2\mu_B(S' - S - |\langle L_z \rangle|/2) \approx 0.77\mu_B$ per formula unit (fmu) would be expected for a single crystal. But measurements on polycrystals [12–14] indicate that $M_s(0)/2$ lies between 0.07 and $0.08\mu_B/\text{fmu}$, corresponding to $M_s(0)$ between 0.14 and $0.16\mu_B/\text{fmu}$. Coronado et al. obtained a remanent magnetization less than $0.03\mu_B$ at 2 K, corresponding to $M_s(0) < 0.06\mu_B$. These results suggest an approximate value of $x_d \approx 0.1$ for the disorder in an $\text{Fe}(\text{II})\text{Fe}(\text{III})$ polycrystal. Such strong disorder may be produced by noncrystallinity or by a deficiency of Fe and the charge-compensating oxidation process $\text{Fe}(\text{II}) \rightarrow \text{Fe}(\text{III})$ [11,12].

We recently proposed a theory [15] for the magnetic order in the $\text{Fe}(\text{II})\text{Fe}(\text{III})$ bimetallic oxalates based on the spin-orbit coupling of the $\text{Fe}(\text{II})$ moments within each bimetallic plane. Our model posited a hierarchy of three energy scales. The Hund's coupling that fixes the spins $S = 2$ and $S' = \frac{5}{2}$ on the $\text{Fe}(\text{II})$ ($3d^6$) and $\text{Fe}(\text{III})$ ($3d^5$) sites is the dominant energy. Next in importance is the C_3 -symmetric crystal-field potential produced by the 6 oxygen atoms that surround each Fe atom. Finally, the contributions to the crystal-field potential that violate C_3 symmetry, the antiferromagnetic exchange coupling $J_c \mathbf{S} \cdot \mathbf{S}'$ between the $\text{Fe}(\text{II})$ and $\text{Fe}(\text{III})$ spins, and the spin-orbit coupling $\lambda \mathbf{L} \cdot \mathbf{S}$ on the $\text{Fe}(\text{II})$ site are all considered to lie in the lowest-energy scale.

The C_3 -symmetric crystal field on the $\text{Fe}(\text{II})$ sites splits the 5-fold degenerate $L = 2$ level into two doublets and one singlet. The expectation value of the orbital angular momentum in the lowest-energy doublet is given by $\pm L_z^{\text{cf}}$, where L_z^{cf} lies between 0 and 2, depending on the

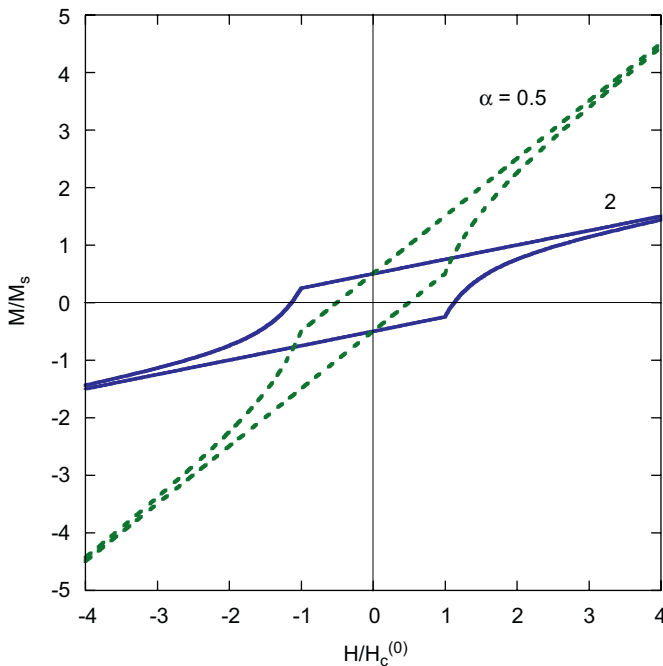


Fig. 1. The magnetization versus field loops for α above and below 1. The coercive field is given by $H_c/H_c^{(0)} = 0.5$ for $\alpha = 0.5$ (dashed) and 1.13 for $\alpha = 2$ (solid).

components of the crystal field. For $|\lambda|/J_c \approx 27$, magnetic compensation occurs when $L_z^{\text{cf}*} < L_z^{\text{cf}} < 1$. By displacing the Fe(II) ions with respect to the oxalate molecules, the cation can shift the value of L_z^{cf} above or below the threshold $L_z^{\text{cf}*} \approx 0.23$ [15].

This model is now used to evaluate the “bare” coercive field $H_c^{(0)}$ for a single-crystal. Our argument follows the same lines as in Ref. [2] except that the anisotropy is now produced by the spin–orbit coupling to a nonquenched orbital angular momentum $\langle \mathbf{L} \rangle$. Consider the domain wall of width $\delta = Na$ lying between $x = -Na/2$ and $Na/2$ sketched in Fig. 2. Here a is the distance between Fe(II) ions along x direction. Because the direction of the magnetic polarization within the domain wall departs from the axis of C_3 symmetry, $\langle \mathbf{L} \rangle$ will be reduced over the whole range Na . Of course, $\langle \mathbf{L} \rangle$ must identically vanish at the center of the domain wall. Hence, the cost of the domain wall in anisotropy energy per unit area is proportional to $|\lambda|N\langle S_z \rangle \langle L_z \rangle / a^2$. Note that $\langle \mathbf{M}_0 \rangle = 2\mu_B \langle \mathbf{S} + \mathbf{S}' + \mathbf{L}/2 \rangle$ will change in magnitude very slightly within the domain wall compared to its bulk value due to the suppression of the orbital contribution. Since the magnetic moment $\langle \mathbf{M}_0 \rangle$ rotates through an angle $\phi = \pi/N$ for each increment $\Delta x = a$, the exchange-energy cost of the domain wall per unit area is proportional to $-J_c \langle S_z \rangle \langle S'_z \rangle \pi^2 / a^2 N > 0$. Keep in mind that $\langle \mathbf{S} \rangle$ and $\langle \mathbf{S}' \rangle$ point in opposite directions.

Therefore, the total energy of the domain wall per unit area can be written

$$\gamma \approx \frac{A}{a^2 N} + \frac{KN}{a^2}, \quad (4)$$

where $A \propto -J_c \langle S_z \rangle \langle S'_z \rangle$ is the exchange energy and $K \propto |\lambda| \langle S_z \rangle \langle L_z \rangle$ is the anisotropy energy. While the exchange energy favors a more gradual change in magnetization and a wider domain wall, the anisotropy energy favors a sharper change in magnetization and a narrower wall. Minimizing γ with respect to N , we find that $N = \sqrt{A/K}$ and $\gamma = 2\sqrt{AK}/a^2$, which are identical to the expressions derived in Ref. [2] when the anisotropy K arises from a single-ion anisotropy term $-D\sum_i S_{iz}^2$ rather than from a nonquenched orbital angular momentum.

The domain-wall energy $\gamma(x)$ depends on position x due to strain that locally perturbs the C_3 -symmetric crystal field, thereby reducing the value of $L_z^{\text{cf}}(x)$. If the strain is written as $\sigma = \sigma_0 \cos(2\pi x/l)$, then $L_z^{\text{cf}}(x)$ and $K(x)$ are

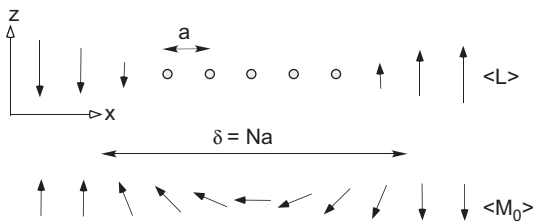


Fig. 2. A domain wall of width $\delta = Na$, showing that the orbital angular momentum $\langle \mathbf{L} \rangle$ becomes very small as the magnetization $\langle \mathbf{M}_0 \rangle$ of the single crystal rotates by 180° . As discussed in Ref. [15], $\langle \mathbf{M}_0 \rangle$ is opposite $\langle \mathbf{L} \rangle$ below T_{comp} .

lowered quadratically as

$$L_z^{\text{cf}}(x) = L_z^{\text{cf}(0)} - \beta\sigma_0^2 \cos^2(2\pi x/l), \quad (5)$$

$$K(x) = K^{(0)} \left\{ 1 - \frac{\beta\sigma_0^2}{L_z^{\text{cf}(0)}} \cos^2(2\pi x/l) \right\}, \quad (6)$$

where $\beta > 0$. For a field applied along \mathbf{z} direction, the instability condition for domain-wall motion is $H_c^{(0)} = a^3(\partial\gamma/\partial x)_{\text{max}}/2M_0$ or

$$H_c^{(0)} = \frac{\pi\Delta K}{M_0}, \quad (7)$$

where $\Delta = (\beta\sigma_0^2/L_z^{\text{cf}(0)})(\delta/l)$ [16]. Since $\beta\sigma_0^2$ is the maximum variation in L_z^{cf} , the first factor in Δ is the relative variation in the orbital angular momentum of the doublet. The second factor, δ/l , gives the ratio of the domain-wall width to the range l of the spatial variation of the strain.

Using $|\lambda|/J_c \approx 27$ and $L_z^{\text{cf}} \approx 0.26$, we estimate that the domain-wall width δ in the Fe(II)Fe(III) bimetallic oxalates is between 1 and 2 lattice constants. Because the domain wall is so narrow, we anticipate that $\delta \ll l$ and $\Delta \ll 1$. Due to the reduced cost in anisotropy energy, domain walls prefer to sit on top of impurities that would otherwise break the local C_3 symmetry. Notice that $H_c^{(0)} \rightarrow 0$ at T_c because K vanishes like the square of the Fe(II) moment. As expected, $H_c^{(0)}$ diverges at the compensation temperature because $M_0 \rightarrow 0$, whereas K remains nonzero at T_{comp} .

We now estimate χ_\perp by assuming that only the Fe(III) moments (with no spin–orbit coupling) respond to a perpendicular magnetic field. Because the Hamiltonian on the Fe(III) sites is given by

$$\mathcal{H}_{\text{III}} = 3J_c \langle S_z \rangle S'_z + 2\mu_B H S'_z, \quad (8)$$

we obtain

$$\chi_\perp = \frac{2\mu_B \langle S'_z \rangle}{H} = -\frac{4\mu_B^2 \langle S'_z \rangle}{3J_c \langle S_z \rangle}. \quad (9)$$

Due to the different temperature dependences of the spins $\langle S_z \rangle$ and $\langle S'_z \rangle$ on the Fe(II) and Fe(III) sites, χ_\perp depends weakly on temperature. At low temperatures, $\tilde{\chi}_\perp \approx 10\mu_B^2/9J_c$, which is about 75% larger than the measured susceptibility [3] if $J_c = 0.45$ meV is estimated using mean-field theory [15]. Since mean-field theory will tend to underestimate J_c , this discrepancy is easily explained.

The coercive field of a polycrystalline sample is evaluated using the formalism of Ref. [15] to obtain $\langle S'_z \rangle$, $\langle S_z \rangle$, and $\langle L_z \rangle$ as functions of temperature. With the experimentally measured $\tilde{\chi}_\perp$ and $x_d = 0.1$ for the disorder, we obtain $\Delta \approx 2.3 \times 10^{-4}$ by fitting the low-temperature coercive field $H_c \approx 0.13$ T measured in Ref. [3]. For $L_z^{\text{cf}} = 0.26$ above the threshold value $L_z^{\text{cf}*} \approx 0.23$ required for magnetic compensation, the coercive field in Fig. 3 vanishes at $T_{\text{comp}} \approx 0.67 T_c$ and has a maximum at roughly $0.6 T_{\text{comp}}$, in agreement with observations of an Fe(II)Fe(III) polycrystal. Since the “bare” coercive field $H_c^{(0)}$ required for

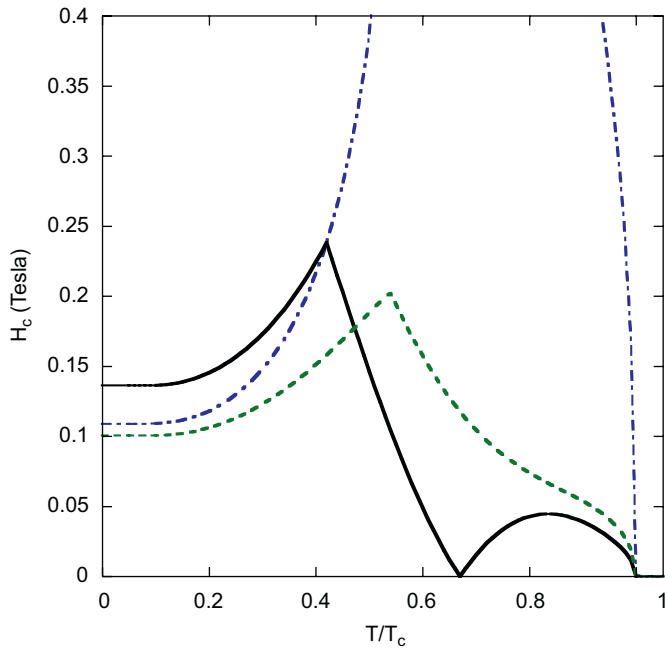


Fig. 3. The coercive field H_c for a polycrystalline bimetallic oxalate with $L_z^{\text{cf}} = 0.26$ (solid) or 0.20 (dashed). The dash-dot curve is the coercive field $H_c^{(0)}$ for a single particle with $L_z^{\text{cf}} = 0.26$ and $T_{\text{comp}}/T_c \approx 0.67$. Other parameters are $|\lambda|/J_c = 27$, $x_d = 0.1$, and $A = 2.3 \times 10^{-4}$.

domain-wall motion still diverges at T_{comp} , the vanishing of H_c at T_{comp} does not imply that domain walls are displaced. In agreement with the general arguments given above, the maximum coercive field satisfies the condition $H_c^{\text{max}} = H_c^{(0)}$.

For $L_z^{\text{cf}} = 0.20 < L_z^{\text{cf}*}$, magnetic compensation is absent. As observed in a $\text{Fe(II)Fe(III)}_{0.9}\text{Cr(III)}_{0.1}$ polycrystal [3] and shown in Fig. 3, H_c^{max} is reduced and shifted to larger temperatures. Each Cr(III) impurity locally breaks the C_3 symmetry about the three neighboring Fe(II) moments. A small number of Cr impurities may actually enhance the coercivity by acting as pinning centers where the domain walls can lower their energy. But a larger fraction of Cr impurities will suppress the coercive field by reducing the overall values of L_z^{cf} and K .

The maximum coercive field $H_c^{\text{max}} \approx 0.23$ T predicted by Fig. 3 is only about 35% of the observed value [3]. This difference probably arises from our over-simplified treatment of the domain-wall energy, which assumed that $\delta/a \gg 1$. The narrow domain wall and strong anisotropy may produce stronger pinning at impurities than our arguments would suggest. In a more realistic description of the polycrystal, the variation in particle sizes will produce distributions in L_z^{cf} and A , smoothing the cusp at H_c^{max} and supporting a nonzero minimum coercivity at T_{comp} .

A test of our model would be to verify that the magnetization is reversible along the straight-line paths in the hysteresis loops of Fig. 1, where the polycrystal is being magnetized perpendicular to the local z -axis of each particle but the domain walls do not actually move. Close

to T_{comp} , the hysteresis loops should be reversible up to large fields.

Polycrystals of other ferrimagnetic materials can potentially also exhibit the predicted behavior. Although an exhaustive search of all candidate materials is beyond our reach, the requirement for both uniaxial anisotropy and magnetic compensation eliminates promising ferrimagnets like the cobaltites [17] (uniaxial anisotropy but no magnetic compensation) and ferrites [18] or rare-earth/transition-metal alloys [19] (magnetic compensation but cubic anisotropy). Materials with a minimum magnetization $M_s(T)$ but no magnetic compensation may not exhibit a minimum coercive field because $\alpha(T) \equiv M_s(T)/(2H_c^{(0)}(T)\tilde{\chi}_{\perp})$ is always greater than 1. So the bimetallic oxalates may provide one of the few examples of this type of behavior.

To conclude, we have shown quite generally that the coercive field of a polycrystalline ferrimagnet with uni-axial anisotropy will be a minimum at the compensation temperature. Those ideas were subsequently applied to the bimetallic oxalates, where such a minimum has been observed [3].

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