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# Finite size and interface mixing effects in Iridium Manganese ultra thin films

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The finite size and temperature dependent properties of antiferromagnets are of critical importance to a wide range of spintronic and neuromorphic computing devices. Here we present atomistic simulations of IrMn, one of the most technologically important antiferromagnets, in both the ordered ( $L1_2$ ) and disordered ( $\gamma$ ) phases. We have found that antiferromagnetic IrMn<sub>3</sub> films show a stronger finite size dependence of the Néel temperature than an equivalent ferromagnet due to the existence of spin frustration. We also find that the disordered  $\gamma$ -IrMn<sub>3</sub> phase shows a dramatic reduction in the Néel temperature to less than room temperature for films less than 1 nm thick. Interfacial intermixing of the IrMn<sub>3</sub> with a non-magnetic Cu capping layer further reduces the Néel temperature for a given film thickness, with a stronger influence on the disordered  $\gamma$ -IrMn<sub>3</sub> phase compared to the ordered  $L1_2$ -IrMn<sub>3</sub> phase. Our results suggest a larger antiferromagnetic film thickness is required for devices operating at or above room temperature compared to an equivalent ferromagnet, particularly for sputtered films with a high degree of interfacial intermixing.

## INTRODUCTION

Antiferromagnetic spintronics[1, 2] is a newly emerging field which has renewed interest in antiferromagnetic (AFM) materials. Antiferromagnets were once thought to be of limited practical use due to their weak interaction with external magnetic fields[3], but when coupled to a ferromagnet to induce exchange bias they find extensive applications in magnetic recording and thermally assisted magnetoresistive random access memory [4, 5]. More recently the discovery of electrical switching of antiferromagnets by staggered spin-orbit torque [6, 7] has led to renewed interest in the fundamental properties of antiferromagnets for spintronics. Antiferromagnets also exhibit ultrafast inertial dynamics due to the balanced nature of magnetic moments at the atomic level which may lead to further improvements in energy efficiency and speed compared to conventional ferromagnetic devices.

Neuromorphic computing[8] is an emerging field which aims to replicate the cognitive capabilities of biological systems and apply them to complex computing problems such as pattern recognition, to study the large scale behavior of cognitive systems and explore new concepts in artificial intelligence. Magnetic devices offer significant potential as artificial neurons due to their intrinsic hysteretic properties [9] and ability to retain their state for extended periods of time. Combining the low power processing capabilities and fast dynamics of antiferromagnetic spintronic devices could enable ultra-low power neuromorphic computing devices. Recently Fukami *et al* [10] developed such a device utilizing exchange bias to preserve its output state while using spin-orbit torque for electrical switching. Once developed, this will allow for devices with can complete complex tasks at high speeds with a low power consumption that can outperform conventional von Neu-

mann computers[8, 11]. Practical spintronic devices rely on interfacial magnetic effects at the atomic scale and so the antiferromagnetic layers have a typical thickness of only a few nanometers. Understanding these effects is particularly important for spin-orbit torque driven devices which rely on interfacial properties for thermal stability and generating torques. Finite size effects in ferromagnets are well understood, but are little studied in antiferromagnets, particularly in materials of practical importance such as Iridium Manganese (IrMn) alloys[12]. IrMn is particularly important due to its high thermal stability[13, 14] and high magnetocrystalline anisotropy energy density[15, 16] and exhibits the largest exchange bias of any known antiferromagnet. Exchange bias provides a unidirectional anisotropy when coupling a ferromagnet (FM) to an antiferromagnet (AFM), causing the FM to be pinned along one direction. The pinning is useful in memory storage and is utilised in many devices such as read heads, spin valves and magnetic sensors. In these devices the AFM plays a supporting role, but their finite size effects are complex. Previously L. Frangou *et al.* found that for films less than 3 nm thick there was a linear decrease of the Néel temperature and for 0.5nm films the Néel temperature falls to only 50K. They note the agreement with the work of D. Petti *et al* [17] who used nanocalorimetry to calculate the Néel temperature of 2 nm thin films of IrMn. The Néel temperature of the 2nm thick film had reduced to 173K, about 20% of the bulk value they calculated. The finite size properties are dependent on the film thickness and ordering and still subject to some interpretation.

Here we present atomistic simulations of the effects of film thickness and interfacial intermixing on the temperature dependent properties of IrMn<sub>3</sub> alloys. We consider both  $L1_2$  ordered and disordered ( $\gamma$ ) phases of IrMn<sub>3</sub> which have different ordering temperatures and thermal stabilities. We have found that there is a weak thickness

dependence of the Néel temperature for films thicker than 2 nm, while for ultra thin films of a few atomic layers we find a catastrophic collapse in the Néel temperature to a few Kelvin. We find that interface mixing is in general a weak effect except at the smallest thickness's where the existence of point defects at the surface leads to large fluctuations of surface spins and a stronger reduction in the Néel temperature for a given film thickness.

### ATOMISTIC SPIN MODEL

To model the magnetic properties of the antiferromagnet (AFM) IrMn we have developed an atomistic spin model with Heisenberg exchange[18]. IrMn alloys have a base face-centred cubic (FCC) unit cell where each of the four sites in the unit cell are magnetically distinct, each comprising a different magnetic sublattice. Here we focus on the technologically relevant IrMn<sub>3</sub> composition considering the ordered L1<sub>2</sub>-IrMn<sub>3</sub> and  $\gamma$ -IrMn<sub>3</sub> phases. In general it is expected that sputtered devices have a relatively low order parameter due to the difficulty in achieving ordered IrMn<sub>3</sub> and so have properties closer to the  $\gamma$ -IrMn<sub>3</sub> phase. In the fully disordered ( $\gamma$ ) phase the Ir atoms are randomly distributed in the four crystal sites within the full crystal lattice. In the fully ordered (L1<sub>2</sub>) phase the Ir atoms are all positioned in the same magnetic sublattice. The model assumes a localized spin density at the Mn atomic sites which forms a fixed length classical spin magnetic moment  $\mu_s = 2.65\mu_B$  calculated from first principles simulations[16]. The Ir sites are treated as non-magnetic impurities that induce a strong spin-orbit coupling effect in the Mn sites. The energy of the system is defined using a spin Hamiltonian of the form:

$$\mathcal{H} = - \sum_{i<j} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j - \frac{k_N}{2} \sum_{i \neq j}^z (\mathbf{S}_i \cdot \mathbf{e}_{ij})^2 \quad (1)$$

where  $i, j$  represent local and neighboring atomic sites and  $\mathbf{S}_i$  and  $\mathbf{S}_j$  the respective spin directions. The effective exchange interactions  $J_{ij}$  were limited to nearest antiferromagnetic ( $J_{ij}^{\text{mn}} = -6.4 \times 10^{-21}$ ) and ferromagnetic next nearest ( $J_{ij}^{\text{mnn}} = 5.1 \times 10^{-21}$ ) neighbors and assumed to be independent of the local atomic ordering. This is justified from first principles simulations which show a similar interatomic range dependence of exchange interactions for the ordered L1<sub>0</sub> and L1<sub>2</sub> phases of IrMn[16]. The magnetocrystalline anisotropy in IrMn<sub>3</sub> is assumed to arise from the large spin-orbit coupling between Mn and Ir sites. Usually the magnetocrystalline anisotropy energy must be determined from first principles *ab-initio* calculations [16]. However, such an approach is particularly

challenging for disordered alloys where the large number of local atomic environments around the Mn sites due to the random placement of Ir in the crystal. We therefore use an approximate yet robust approach using the Néel pair anisotropy model [3, 19] where  $\mathbf{S}_i$  and  $\mathbf{S}_j$  are unit vectors describing the spin direction at the local site  $i$  and neighboring  $j$  sites respectively.  $k_{ij} = 4.22 \times 10^{-22}$  is the Néel pair anisotropy constant between Mn and Ir sites and  $\mathbf{e}_{ij}$  is a unit position vector between nearest neighboring Mn and Ir sites.

The temperature dependent properties of the system are simulated using a metropolis Monte Carlo algorithm using the Hinzke-Nowak update algorithm[20] using a combination of spin-flip, random and Gaussian trial spin moves. Each sample 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 1 000 000 Monte Carlo steps to find a ground state spin configuration. The system was then heated back up to 1500 K to find the Néel temperature in steps of 10 K performing 10 000 equilibrating Monte Carlo steps and then averaging over a further 10 000 Monte Carlo steps. All numerical simulations were performed using the VAMPIRE software package[18, 21].

The Néel temperature is determined from the mean sublattice magnetization  $n$  which is the average magnetization length calculated from a summation over the spins of each distinct magnetic sublattice. As a classical simulation the temperature dependent magnetization is accurately described by the fitting function

$$n(T) = n_0 \left( 1 - \frac{T}{T_N} \right)^\beta \quad (2)$$

where  $n$  is the average sublattice magnetization,  $n_0$  is the average zero-temperature ordering of the sublattices,  $T$  is the temperature,  $T_N$  is the Neel temperature and  $\beta$  is the critical magnetization exponent. It is possible calculate the Néel temperature from the sublattice magnetization against temperature curve. However, for small sizes the fitting becomes inaccurate due to a reduced criticality of the magnetization, and so we extract the Néel temperature from the peak in the isotropic longitudinal susceptibility  $\chi_n$  for each sublattice given by

$$\chi_n = \frac{\sum_i \mu_i}{k_B T} (\langle |n|^2 \rangle - \langle n \rangle^2) \quad (3)$$

where  $i$  are indices of atoms within the same sublattice. Fig. 1 shows a typical simulation result for a (10 nm)<sup>3</sup> cube of L1<sub>2</sub> IrMn<sub>3</sub> showing the usual decrease in sublattice spin order with increasing temperature due to spin fluctuations. The sublattice susceptibility diverges

at the Néel temperature with a well defined peak from which we extract the  $T_N$ .

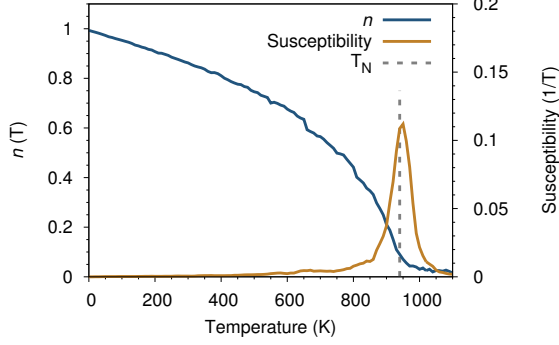


FIG. 1. Plot of the the average sublattice magnetization  $n$  and isotropic longitudinal susceptibility  $\chi_n$  as a function of temperature for the  $L1_2$  phase of  $\text{IrMn}_3$ . The Néel temperature  $T_N$  is extracted from the peak in the susceptibility and is close to the bulk value[14] of 1000 K.

### BULK MAGNETIC PROPERTIES OF IRIIDIUM MANGANESE

To validate our model we have simulated the ground state spin structures of the ordered  $L1_2$  and disordered  $\gamma$  phases of  $\text{IrMn}_3$  as shown in Fig. 2 using the Monte Carlo Metropolis algorithm by zero-field cooling from above the Néel temperature to 0 K. In agreement with previous experimental [13, 14] and *ab-initio* results [16] we find ordered  $L1_2$ - $\text{IrMn}_3$  has a triangular ( $T1$ ) spin structure where the magnetic moments lie in plane along the  $[111]$  planes with an angle of  $120^\circ$  degrees between them pointing along the  $[211]$  directions and that disordered  $\gamma$ - $\text{IrMn}_3$  has a tetrahedral ( $Q3$ ) spin structure[13] where the magnetic moments point  $109.5^\circ$  apart.

Our simulations reproduce the different ordering temperatures of the  $L1_2$  ( $T_N \sim 1000$  K) and  $\gamma$  ( $T_N \sim 700$  K) despite using the same Spin Hamiltonian and both ordered  $L1_2$ - $\text{IrMn}_3$  and disordered  $\gamma$ - $\text{IrMn}_3$  having the same average number of exchange bonds. This arises due to different degrees of geometric frustration in the spin structures. All Mn spins in the system energetically prefer full antiferromagnetic alignment ( $180^\circ$ ), but due to the geometric arrangement and high coordination numbers this spin structure is not possible. Therefore, in the ground state the spins are already frustrated and so thermal spin fluctuations have a stronger effect on the sublattice ordering in the  $\gamma$  phase compared to the  $L1_2$  phase. This naturally leads to a lower ordering temperature in the  $\gamma$ - $\text{IrMn}_3$  phase.

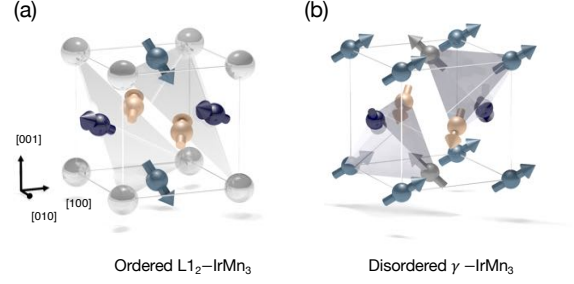


FIG. 2. Visualization of the simulated ground state spin structures of  $\text{IrMn}_3$  obtained from zero-field cooling. The spins show an average spin of each magnetic sublattice direction over the whole sample. In the case of  $L1_2$ - $\text{IrMn}_3$  the corner atoms are all Ir and so have no net magnetic moment. The spin structures are in excellent agreement with experimental measurements and first principles simulations.

### FINITE SIZE EFFECTS IN ULTRATHIN IRMN FILMS

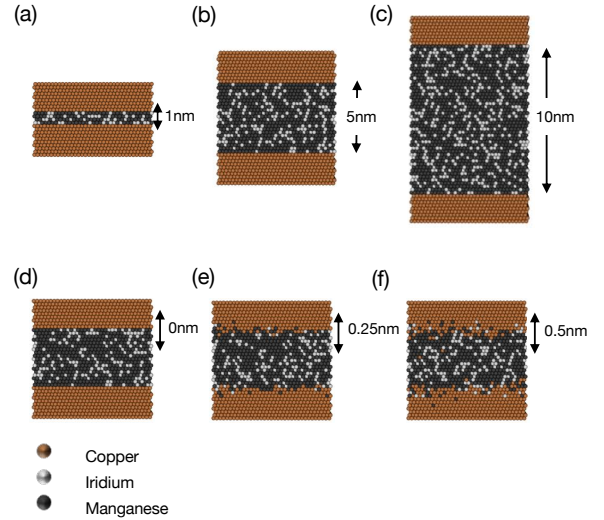


FIG. 3. Atomistic visualization of the generated thin film system for the  $\gamma\text{IrMn}_3$  phase for different film thicknesses (a) 1 nm, (b) 4 nm and (c) 10 nm. The colors represent different atomic species. The same is shown for different levels of interface mixing with interface widths of (d) 0 nm, (e) 0.25 nm and (f) 0.5 nm.

The properties of ultrathin films of  $\text{IrMn}_3$  were modeled assuming the layer is sandwiched between non-magnetic Cu layers with low spin-orbit coupling so that they have no influence on the magnetic parameters other than the loss of Mn exchange bonds at the surface of the film. The simulated system has fixed lateral dimensions of  $15 \text{ nm} \times 15 \text{ nm}$  with periodic boundary conditions in the plane and a varying thickness in the range 0.25 – 10

nm. A visualization of the generated crystal structure for the disordered  $\gamma$  phase is shown in Fig. 3. Initially the interface between the Cu and IrMn<sub>3</sub> is assumed to be atomically flat but we also study the case of intermixing between the layers to recreate the effects of sputtered films where atomic mixing is more likely to occur. The mixed interface was generated using a probability distribution defined by

$$P(z) = 1 - \frac{1}{2} \tanh\left(\frac{\pi(z - z_0)}{w}\right) \quad (4)$$

where  $P(z)$  is the probability of finding an atom of a particular type at height  $z$ ,  $z_0$  is the interface height and  $w$  is the width of the interface. Every atom in the IrMn layer is initially set as Ir or Mn and then has a probability ( $P$ ) of being changed to a Cu atom depending on its height ( $z$ ). The reverse process is done for the mixing of Ir and Mn into the Cu layers. The width of the interface was systematically varied between 0 and 0.5 nm as shown schematically in Fig 3.

The temperature dependent sublattice magnetization and isotropic susceptibility was calculated for varying thicknesses of IrMn<sub>3</sub>. Due to the large scatter in the calculated properties in the lowest film thicknesses the simulations are averaged over 10 different sample crystal structures with different pseudorandom number sequences in the Monte Carlo algorithm to obtain average data for the mean sublattice magnetization and susceptibility. A typical set of data for thickness  $t_{\text{IrMn}} = 1$  nm and atomically flat interface  $w = 0$  nm is shown in Fig. 4 comparing the (a) disordered  $\gamma$  and (b) L1<sub>2</sub> phases. The simulations show a significant decrease in the Néel temperature for both phases due to the missing interface Mn-Mn exchange bonds. In particular the  $\gamma$  phase shows a Néel temperature close to room temperature even for an atomically smooth interface, suggesting that films thinner than this are unsuitable for applications in spintronics.

Interestingly the zero temperature ordering of the disordered  $\gamma$  phase obtained from the field cooling simulation is significantly less than 1, with a global ordering fraction around 0.7. The small size of the system means that the spins should form a single antiferromagnetic domain. However, the random distribution of the Ir sites in the crystal means that some areas of the AFM are weakly coupled to the rest allowing a stable antiferromagnetic domain to form. This effect is unique to thin films, since in the bulk there is a path which allows the exchange coupling of regions around Ir-rich regions of low coupling, while in very thin films the percolation route in one dimension is constrained and leads to decoupling of different regions of the crystal. This suggests that even in the absence of thermal fluctuations, full ordering of the IrMn<sub>3</sub> at nanoscale sizes may be difficult to achieve.

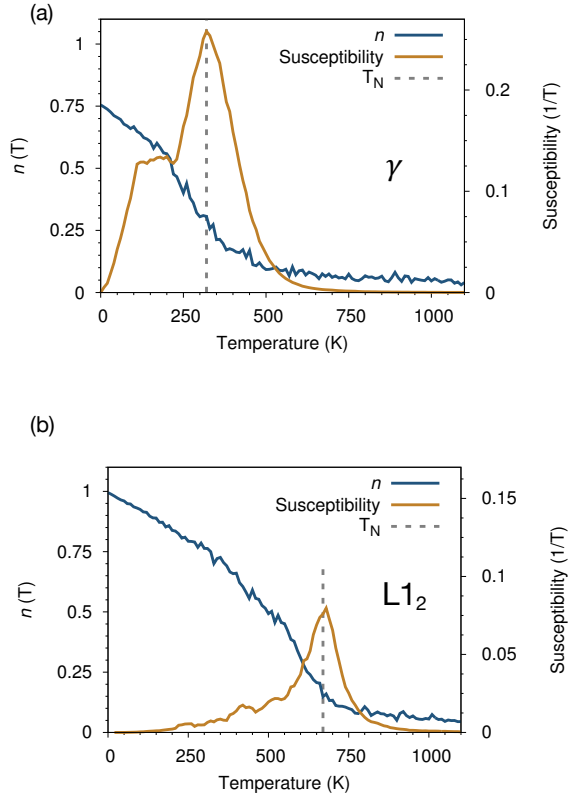


FIG. 4. Simulated temperature dependent sublattice magnetization curves and isotropic susceptibility for a 1 nm thick thin film of IrMn<sub>3</sub> comparing  $\gamma$  (a) and L1<sub>2</sub> (b) phases for a system with a perfectly flat interface. The data are averaged over ten statistically independent samples. The points show simulated data and the line is a fit using Eq. 2. Both curves show a significant reduction in the Néel temperature compared to bulk and reduced criticality near  $T_N$  due to the small finite thickness of the film.

A side effect of the intrinsically low magnetic order is a discontinuity in the isotropic susceptibility at very low temperatures. Here, infinitesimal thermal fluctuations of the spins lead to a large variation of the sublattice magnetization, in strong contrast to the fully ordered L1<sub>2</sub> phase which has a low susceptibility at low temperatures. This is an unusual effect and may be measurable as an intrinsic instability of the antiferromagnetic spin structure.

A systematic study of the effect of film thickness and intermixing on the Néel temperature is shown in Fig. 5 for the  $\gamma$  (a) and L1<sub>2</sub> (b) phases alongside comparative simulation data for a generic FCC ferromagnet with a Curie temperature the same as the Néel temperature. Both antiferromagnetic phases show a stronger finite size effect than the comparable ferromagnetic film, with a stronger reduction in the ordering (Néel) temperature

for a given film thickness. This is likely due to the geometric spin frustration which increases the influence of the thermal spin fluctuations on the sublattice ordering. Comparing the two phases of IrMn<sub>3</sub>, the  $\gamma$  phase shows a stronger finite size effect and larger reduction in the Néel temperature compared to the ordered phase. We attribute this to the reduction in global sublattice magnetic order in the  $\gamma$  phase due to the formation of antiferromagnetic domains and larger effect of thermal fluctuations. For the thinnest films there is a catastrophic reduction of the Néel temperature to a few degrees Kelvin which is not seen in the ferromagnetic case. Here we believe that the origin is due to the percolation effect where locally antiferromagnetic order still exists, but even on the nanoscale the long range order is disrupted leading to an apparent low Néel temperature over the total simulated sample. These effects may be more severe for laterally larger films which would typically be used in devices.

Finally we consider the effect of intermixing to emulate the effect of sputtered thin films as shown by the comparative data in Fig. 5. It is clear from the data that larger interfacial mixing leads to a larger reduction of the Néel temperature of the films compared to the atomically smooth interface. The effect also seems to be larger for the  $\gamma$  phase compared to the ordered L1<sub>2</sub> phase, likely due to the random intermixing increasing the exchange decoupling of different regions of the antiferromagnet and reducing the global ordering. For the thicker films  $t_{\text{IrMn}} > 3$  nm the effects of intermixing on the Néel temperature is much weaker and likely negligible for devices operating close to room temperature.

## DISCUSSION

We have investigated the effect of film thickness and intermixing on the temperature dependent properties of antiferromagnetic IrMn<sub>3</sub> alloys using an atomistic spin model and Monte Carlo Metropolis simulations. The temperature dependent magnetic properties of the ordered L1<sub>2</sub> and disordered  $\gamma$  phases of IrMn<sub>3</sub> have been calculated for ultra thin films from which we extract the sublattice susceptibility and Néel ordering temperature. We find a relatively strong finite size effect compared to a ferromagnetic system arising due to spin frustration and a larger influence of thermal spin fluctuations on the magnetic order. For the thinnest  $\gamma$ -IrMn<sub>3</sub> films we find a collapse of the Néel temperature to less than room temperature making them unsuitable for devices due to their large thermal spin fluctuations and instability. In general moderate intermixing has a weak effect for films thicker than 3 nm, but causes a severe reduction in the Néel temperature for the thinnest films. We conclude that more control of the film properties and the use of thicker

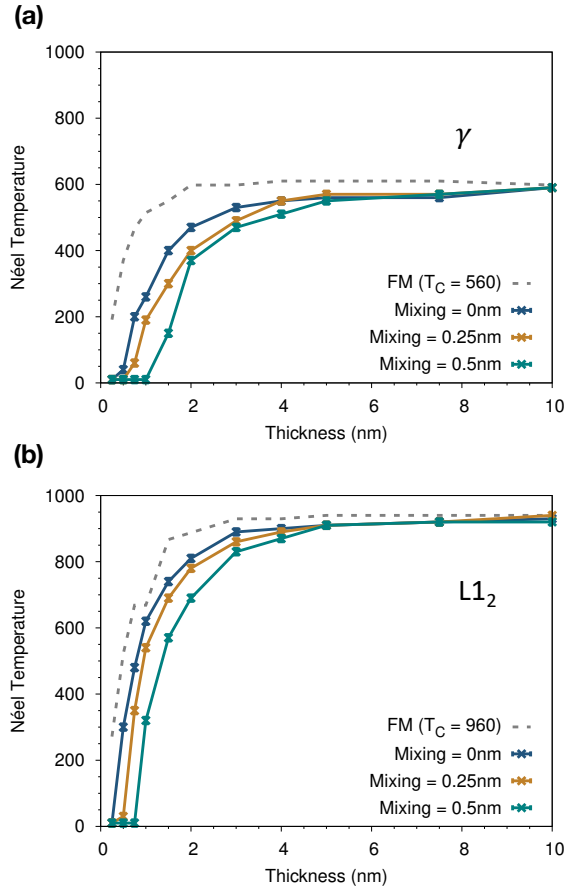


FIG. 5. Simulated systematic variation in the Néel temperature with varying film thickness for IrMn in both the  $\gamma$  (a) and L1<sub>2</sub> phases (b) for different interfacial mixing widths. The dashed lines show comparative data for a ferromagnet with the Curie temperature  $T_C = T_N$  for each phase. The data show a systematic decrease in the Néel temperature for thinner films, and a stronger finite size effect in the  $\gamma$  phase compared to the L1<sub>2</sub> phase. In both cases the finite size variation of the Néel temperature shows a stronger decrease than the equivalent ferromagnet (FM) due to the inherent spin disorder. Intermixing of the IrMn with a non-magnetic Cu layer shows an enhanced finite size effect for increased mixing due to a larger number of missing exchange bonds.

layers is required for devices utilizing antiferromagnetic IrMn films compared to equivalent ferromagnetic thin films. Although the results from our study are specific to IrMn<sub>3</sub>, we expect that other antiferromagnets with a similarly complex spin structure will also show strong finite size effects, for example PtMn and FeMn. Simple two-sublattice antiferromagnets such as CoO, NiO, CuMnAs and Mn<sub>2</sub>Au conversely have a lower degree of spin frustration and so may not be as susceptible to finite size effects, and therefore may be better suited to applications.

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