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Are half-metallic ferromagnets half metals? (invited)

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Several classes of materials are currently under investigation as potential high-spin-polarization materials. Unfortunately, the proposed half-metallic materials, including the semi-Heusler alloys, the manganese perovskites, and the “simpler” oxides such as chromium dioxide and magnetite, suffer from fundamental limitations. First, the postulated half-metallic systems lose their full ($T = 0$) spin polarization at finite temperatures and, second, surfaces, interfaces, and structural inhomogeneities destroy the complete spin polarization of half-metallic systems even at zero temperature. In a strict sense, half-metallic ferromagnetism is limited to zero temperature since magnon and phonon effects lead to reductions in polarization at finite temperatures. © 2004 American Institute of Physics. [DOI: 10.1063/1.1682911]

I. INTRODUCTION

By definition, the half-metallic ferromagnets have only one spin channel for conduction at the Fermi energy and thus are potentially of considerable interest in spin electronics.^{1–3} The proposal that there might exist such half-metallic ferromagnetic materials with 100% zero-temperature spin polarization at the Fermi level has quite a long history^{4–8} dating back to de Groot.^{5,6,8} Among the most cited candidates for 100% spin polarization are the semi-Heusler alloys (NiMnSb),^{5–12} “full” Heusler alloys,^{12–14} zinc-blende structure materials,^{15–17} colossal magnetoresistance materials [$\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$,^{18,19} $\text{Sr}_2\text{FeMoO}_6$ (Ref. 20)], and semimetallic magnetic oxides [CrO_2 (Refs. 21–24) and Fe_3O_4 (Refs. 25 and 26)]. A number of other materials have also been suggested as half-metallic ferromagnets.^{4,27}

Practical electronic devices that exploit the spin as well as the charge of the electron,^{28,29} including nonvolatile magnetic random-access memory elements,^{29,30} operate at non-zero temperatures, ideally at or above room temperature. Unfortunately, many potential half-metallic systems exhibit dramatic decreases in spin polarization and junction magnetoresistance well below room temperature, as discussed below. It is, therefore, necessary to consider finite temperature effects such as magnon and phonon excitations. Furthermore, if spin injection into a semiconductor, or spin-polarized tunneling is the goal, then the properties of the interface have to be considered, including the effects of finite temperatures on the electronic, magnonic, and phononic states at the interface and the quality of the interface. As we will elaborate (*vide infra*), these complications are common to all potential half-metallic systems, so that the question arises whether these materials are indeed “operationally” better spin injectors than Fe (40%), Co (34%), or even Ni (23%).

To understand the effect of thermal excitations and reduced symmetries at surfaces and defects, one must take into account the localized features of the iron-series $3d$ electrons, even in itinerant systems. In itinerant magnets, the $3d$ electrons hop from atom to atom, and therefore contribute to the conductivity, but during their temporary residence on a given

atomic site they adapt to the local environment. This is seen most clearly at finite temperatures,³¹ where spin disorder can be approximated by random intra-atomic exchange fields.^{32,33} Essentially, the effect of local disorder is to rotate the local spin direction and thus modify the local magnetic moment³⁴ and spin polarization.^{35,36} The coupling between the atomic moments can then be understood in terms of Heisenberg-type exchange constants, and the signs and magnitudes of the constants determine, for example, whether the spin structure is collinear or noncollinear. In addition, there is a small³⁴ noncollinearity due to spin-orbit coupling.^{35–38}

The modification of the spin structures due to local perturbations is a relatively common phenomenon and easily rationalized in terms of a Landau-Ginzburg approach.^{34,39,40} The idea is to treat the atoms as a continuum in which exchange interactions are described by a gradient term. For example, at surfaces the approach yields an exponentially decaying perturbation.^{39,40} Surfaces, interfaces, defects, thermal disorder, and other aperiodicities have a pronounced effect on the spin polarization.

II. SPIN DISORDER AND CONDUCTIVITY

As noted now by several groups,^{41–44} spin disorder has a profound influence on the persistence of half-metallic character at finite temperature. Spin-mixing effects in ferromagnetic metals have been investigated for many years, for example, in the context of finite-temperature excitations in elemental nickel.³² A simple tight-binding description is provided by

$$H_{\mu\nu ik\sigma\sigma'} = T_{\mu\nu ik} \delta_{\alpha\beta} - V_{\mu} \delta_{\mu\nu} \delta_{ik} \delta_{\alpha\beta} - I_o \delta_{\mu\nu} \delta_{ik} \mathbf{e}_i \cdot \boldsymbol{\sigma}_{\alpha\beta}, \quad (1)$$

where $T_{\mu\nu ik} = \langle \phi_{\mu}(\mathbf{r} - \mathbf{R}_i) | T | \phi_{\nu}(\mathbf{r} - \mathbf{R}_k) \rangle$ is the hopping integral between a μ -type orbital at \mathbf{R}_i and a ν -type orbital at \mathbf{R}_k , V_{μ} is the crystal-field energy for the μ -type orbital, I_o is the intra-atomic d -electron exchange, and \mathbf{e}_i is the local magnetization direction. In terms of Eq. (1), ferromagnetism results from the I_o Stoner term, which reduces and enhances the energies of \uparrow and \downarrow electrons, respectively.

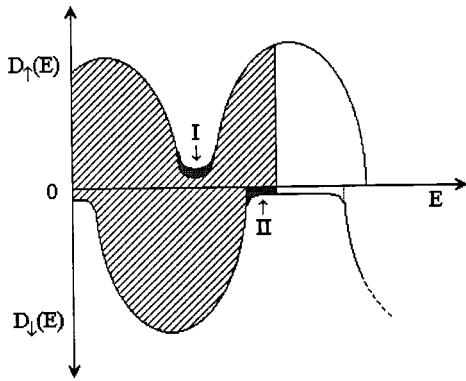


FIG. 1. Schematic \uparrow and \downarrow densities of states (dark shadowing denotes regions occupied at finite temperature only).

We have used Eq. (1) to model half-metallic ferromagnets^{41,42} and to compare with ordinary itinerant ferromagnets. As in other systems,^{32,33} the description of finite-temperature spin disorder involves the standard spin-1/2 rotation matrix $\hat{U}(\phi, \theta)$, which describes the rotation of a \uparrow spin by the angles θ and ϕ . The inverse matrix $\hat{U}^{-1} = \hat{U}^+(\phi_i, \theta_i)$ diagonalizes the exchange term $\mathbf{e}_i \cdot \boldsymbol{\sigma}_{\alpha\beta}$ in Eq. (1), while applying the unitary matrix $\Pi_i \hat{U}^+(\phi_i, \theta_i)$ to Eq. (1) yields

$$H_{\mu\nu i k \alpha \beta}^i = T_{\mu\nu i k} \sum_{\gamma} U_{\alpha\gamma}(\phi_i, \theta_i) U_{\gamma\beta}^+(\phi_k, \theta_k) - (V_{\mu} \delta_{\alpha\beta} + I_o \sigma_{z\alpha\beta}) \delta_{\mu\nu} \delta_{ik}, \quad (2)$$

where the thermal spin disorder has been mapped onto random hopping integrals.

Far below T_c , the densities of states (DOS) of ordinary ferromagnets are only weakly modified by thermal fluctuations, because $k_B T$ is much smaller than the Fermi energy. However, the behavior of half-metallic ferromagnets cannot be reduced to a smearing of the Fermi level by a relative amount of the order of $k_B T / E_F$.⁴¹⁻⁴⁴ Figure 1 shows typical finite-temperature \uparrow and \downarrow densities of state.^{41,42} The distortion of the \uparrow DOS, $D_{\uparrow}(E)$, has the character of a quantitative correction (dark region I), but in the \downarrow gap the DOS changes qualitatively, from zero to nonzero (dark region II). This spin mixing yields nonzero $D_{\downarrow}(E)$ values in the middle of the spin-down gap, well above the highest energy level occupied at zero temperature. As a consequence, the resistance of the \downarrow channel changes from infinity to a finite value.

The appearance of the spin-down density (II) in Fig. 1 has a simple physical interpretation: a \uparrow electron hops onto a neighboring atomic site characterized by a different axis of spin quantization. In the frame of the neighboring atom, the spin wave function has nonzero projections onto both \uparrow and \downarrow states. For random disorder, the spin mixing can be interpreted as an Anderson localization of spin-polarized $3d$ electrons in a thermally randomized atomic potential. In a very crude approximation, the magnitude of this spin-mixing contribution to the DOS is proportional to $M_o - M_s(T)$, where $M_s(T)$ is the spontaneous magnetization and $M_o = M_s(0)$.^{41,42} Considering a ferromagnet-insulator-ferromagnet tunnel-junction geometry, Itoh, Ohsawa, and Inoue⁴³ have calculated the spin polarization at interfaces

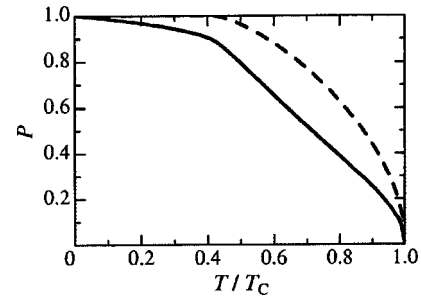


FIG. 2. The temperature dependence of the polarization P , either with (solid) and without (dashed) spin fluctuations for a half metal, adapted from Ref. 43.

with and without spin fluctuations considered. Figure 2 indicates that the effect of spin fluctuations is quite significant.

In Sec. III, we will see that the many-sublattice character of typical half metals modifies this picture. There is enhanced magnon and phonon coupling, with a suppression of 100% spin polarization at the Fermi level.

III. MAGNONS AND PHONONS

Magnon excitations lead to small decreases in polarization at low temperatures, more or less proportional to the drop in magnetization, but dramatic drops in polarization have been observed well below room temperature for many potential half-metallic systems. The tunnel magnetoresistance of $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3/\text{SrTiO}_3/\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ where $x = 0.2$ and $x = 0.3$ dramatically drops at about 27 K (Ref. 45) and 130 K (Refs. 46–50), respectively. Dramatic drops in the measured polarization have been seen for $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$,⁵¹ CrO_2 ,^{52,53} and NiMnSb .⁵⁴⁻⁵⁷ These decreases in polarization suggest that yet another temperature dependent mechanism for depolarization exists.

Gauging the magnitude of the spin polarization at the Fermi level and its pronounced drop well below room temperature, we must take into account the multisublattice character of potential half-metallic ferromagnets.^{41,42} For example, increasing the temperature of $\text{NiMnSb}(100)$ beyond 80 K leads to pronounced changes in the band structure polarization, as probed by magnetic circular dichroism (MCD),⁵⁴ shown in Fig. 3 and summarized in Fig. 4. This is accompanied by a decrease in the $\langle 200 \rangle$ neutron magnetic scattering factor with increasing temperature^{54,55} (inset of Fig. 4). This as well as both the differential magnetoresistance $[d(\Delta R/R)/d(\mu_0 H)]$ [Fig. 4(a)] and the resistance [Fig. 4(b)] of NiMnSb are consistent with a crossover in magnetic ordering at 80 to 100 K. At temperatures above this metamagnetic transition, all indications suggest an onset of a large spin minority channel.⁵⁴

Associated with the compound structure of all the postulated half metals, there are typically a number of low-energy transverse and longitudinal optical modes. These optical phonon modes can couple to spin-wave modes and reduce the net magnetization.⁴² In real space, these magnetization modes correspond to a tilting between neighboring atomic spins, thus the tilt angle depends on the k vector where the phonon and magnon bands cross. Generally, the

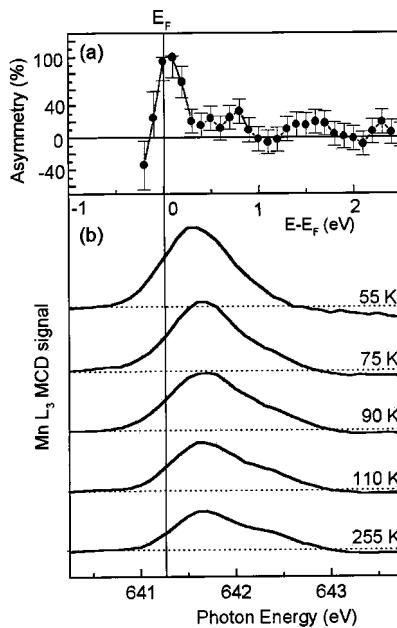


FIG. 3. The polarization asymmetry for NiMnSb(100) from spin-polarized inverse photoemission at the elevated temperature of 300 K (a) is compared to the temperature dependent magnetic circular dichroism signal data across Mn 2*p* (b) core levels (Ref. 54). The MCD signal is shown for several different temperatures, increasing as the temperature decreases.

closer to the Brillouin zone edge the magnons and transverse optical modes couple, the greater the tilt angle. The corresponding moment misalignment gives rise to a pronounced spin mixing.

In the case of NiMnSb, nearly dispersionless transverse and longitudinal optical modes are observed at about 28

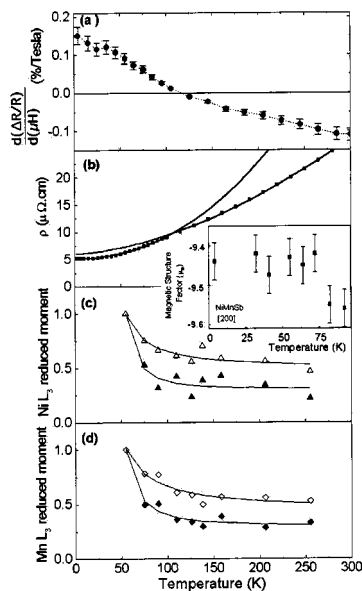


FIG. 4. Evidence of the metamagnetic transition in NiMnSb, taken from (Ref. 54), with permission. (a) Magnetoresistance slope [$d(\Delta R/R)/d(\mu_0 H)$], (b) the resistivity, (c) the relative Ni moment, and (d) relative Mn moment derived from MCD are compared. The change in the magnetic structure factor for the $\langle 200 \rangle$ peak in neutron scattering shown as an inset, as adapted from (Ref. 55). The T^2 and $T^{-1.65}$ curves are fitted to the resistance data.

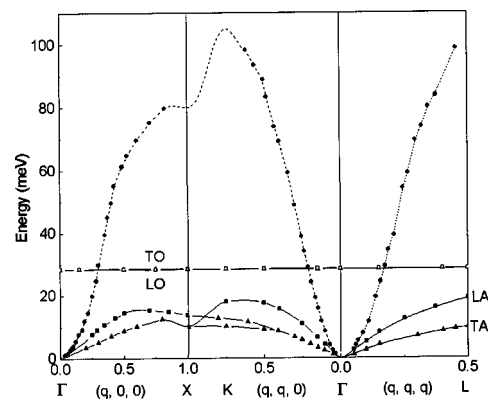


FIG. 5. The phonon and magnon band structure for NiMnSb taken from (Ref. 58). Dispersion curves for magnons (●), longitudinal phonons (■), and transversal phonons (▲) at 300 K are shown. No magnetic field was applied.

meV,⁵⁸ as illustrated by the experimental phonon band structure reproduced in Fig. 5. Magnon coupling to these phonon modes should result a dramatic loss in Ni and Mn magnetization and polarization in NiMnSb, well below room temperature, as is observed.⁵⁴ In the semi-Heusler alloys, this corresponds to a tilting between neighboring Ni and Mn atoms. In real space, the coupled magnon-phonon mode may cant the spins between Mn next nearest neighbors to form an angle of the order of 60°, while all the Mn moments are misaligned with respect to the nickel moments.⁵⁴

Manganese perovskites exhibit a low-temperature spin-wave softening and broadening of magnetoelastic origin, closely related to the proximity of the charge-ordered insulating state.⁵⁹ The effect occurs where the magnon dispersion crosses the longitudinal optical branch of the phonons, at $ka/2\pi=0.3$.^{59,60} The lowest transverse optical modes in these systems, about 23 meV for $\text{La}_{0.7}\text{A}_{0.3}\text{MnO}_3$ ($A = \text{Ca}, \text{Sr}$),^{59,60} suggest metamagnetic transitions should occur in the manganese perovskites, similar to those observed in NiMnSb,⁵⁸ at about 84 K. This suggests that the loss of net spin polarization should occur, and indeed there are indications this does occur, as noted above, at about 100–130 K.^{46–50} The magnon phonon coupling is quite strong,^{61,62} leading to complete damping of the magnons from about $ka/2\pi \approx 0.3$ to the Brillouin zone boundary.^{59–61} Similar phonon modes exist in a variety of compounds, including materials such as CrO_2 and Fe_3O_4 . That phonon effects should be so profound should not be much of a surprise, given the existence of a charge ordering transition in the perovskites and the nonmetal to metal Verwey transition in Fe_3O_4 .

Note that an elevated Curie temperature T_c is *not* a good indicator of spin polarization at elevated temperatures. Semi-Heusler alloys ABC ($A = \text{Ni}, \text{Pd}, \text{Pt}$; $B = \text{Mn}$; $C = \text{P}, \text{As}, \text{Sb}$) with a T_c of about 740 K have just as significant drops in polarization below room temperature as the manganese perovskites: $\text{La}_{0.35}\text{A}_{0.65}\text{MnO}_3$ ($A = \text{Sr}, \text{Ba}, \text{Pb}$) with a $T_c = 360$ K or CrO_2 with a $T_c = 397$ K. First, from other many-sublattice magnets,⁶³ it is known that different sublattice magnetizations have different temperature dependencies. In

small magnetic fields, of interest in the present context, this effect may be very pronounced, but high magnetic fields tend to align sublattices and smooth noncollinearities. This enhances the magnetization well below T_c . In extreme cases, the zero magnetic field magnetization curve $M(T)$ should indicate a magnetization far smaller than the magnetization curves for the same sample taken with moderate to high magnetic fields, for much of the temperature region below the ferromagnetic Curie temperature. A similar effect is encountered at interfaces.⁴³ Second, different transition-metal sublattices yield, in general, different spin-scattering contributions per atom.

IV. THE PROBLEM OF INTERFACES

It is very difficult to see how magnons can be eliminated from potential half-metallic systems to provide true half-metal behavior. One could consider suppressing the long wavelength (and low-energy) magnons by breaking up the half-metallic ferromagnetic crystal into small crystallites, but this approach does not provide any advantage as even stronger spin-mixing effects are caused by *surfaces* and *interfaces*. Spin minority surface states, well known in half metals,^{8,12,17,64–66} can “develop” into interface states^{65–67} and therefore lead to the loss of half-metallic character. Indeed the creation of more interfaces presents a considerable problem as increases in interface density result in increases in the spin minority population decreasing the overall spin polarization as well, even near 0 K.

For the stoichiometric surface, a surface electronic structure that differs from the bulk should not be surprising because there is experimental evidence of large surface enthalpy differences from the bulk in these materials. There are strong chemical potential for surface segregation in these materials^{68,69} indicating that the equilibrium surface is not the same stoichiometry as the bulk.^{68–73}

V. EXPERIMENTAL PROOF OF HALF-METALLIC FERROMAGNETISM

There have been various experimental arguments in favor of true half-metallic ferromagnetism.^{1,2,4,46,74–82} Polarization measurements at very low temperature^{46,80–82} do not yield compelling evidence that there are no magnon contributions at low temperatures, even at 1.6 K. For example, even though tunnel junctions include contributions from the spin dependent Fermi velocity, which could effectively enhance the tunnel magnetoresistance,⁸³ no tunnel magnetoresistive junction measurement has provided a strong indication of half-metallic character. The highest tunnel-magnetoresistance values reached have been as high as 150% to 730% (Refs. 45, 47, 48, and 51) to 1800%,⁴⁶ and this only at very low temperatures (though very high values of magnetoresistance have been observed at 77 K,⁵¹ and room temperature⁸⁴). Unfortunately, interface and defect scattering contributions to the magnetoresistance are difficult to eliminate, making such measurements less than compelling. It is hard to see how magnon scattering can be eliminated from magnetoresistive junctions at small applied bias.⁸⁵ Unfortunately, indirect proof of half-metallic character from the

TABLE I. Experimental spin polarization determined by spin-polarized electron spectroscopies at elevated sample temperatures. With the exception of the spin-polarized *inverse* photoemission experiments in (Refs. 54, 71, 73, and 78), all data were obtained by spin-polarized photoemission.

Potential half-metallic system	Polarization claim	Temperature (K)
La _{0.7} Sr _{0.3} MnO ₃	100±5% (Ref. 74)	40
	100% (Ref. 78)	100
La _{0.7} Pb _{0.3} MnO ₃	Much less than 80% (Ref. 71)	300
	13% (Ref. 100)	10
CrO ₂	95% (Ref. 75)	293
	95% (Ref. 76)	293
Fe ₃ O ₄	−80±5% (Ref. 77)	293
	−40% (Ref. 91)	130
NiMnSb	50% (Ref. 92)	300
	40% (Ref. 93)	300
	High but not near 100% (Refs. 54 and 73)	300

magnetic moment measured at high fields is also likely to be very insensitive to a small spin minority density of states.

Transport spin-polarization measurements, using point-contact Andreev reflection, have generally provided the highest measured polarizations to date.^{80–82,86–90} In spite of several claims of proof of half-metallic character for CrO₂, from point contact Andreev reflection,^{80,81} superconducting tunnel junctions⁸² and magneto-resistive transport⁵² measurements, these measured values of polarization (96±1%,⁸⁰ 98.4%,⁸¹ 97%,⁸² 82%,⁵² respectively) generally miss the mark of 100% polarization. The analysis of the data must be taken with considerable care^{86,89} and cannot ignore Fermi velocity contributions. In the case of La_{2/3}Sr_{1/3}MnO₃, the appreciable spin minority population at 1.5 K is probably a band-structure effect.⁸⁶ High polarization combined with a large Fermi velocity in one spin channel, versus low polarization and large localization in the other spin channel, can accentuate an already high polarization in many transport measurements.

There have also been claims of 100% spin polarization at temperature well above zero kelvin. Apart from a somewhat ambiguous scanning tunneling microscopy spectroscopy study undertaken at 77 K,⁷⁹ claims of 100% spin polarization at more elevated temperatures (>40 K) rest on evidence from spin-polarized photoemission^{75–77} and spin-polarized inverse photoemission,⁷⁸ as summarized in Table I. Spin-resolved photoemission measurements^{75–77} and spin-polarized inverse photoemission⁷⁸ that claim to provide evidence of half-metal behavior, in fact, do not. (Of course, not all such measurements of potential half-metallic systems have led to such claims.^{71,73,91–93}) Finite temperature effects leading to the population of spin minority states near E_F , as outlined in the previous sections, would be most significant at wave vectors away from $\bar{\Gamma}$. For this reason, they may not be observed by spin-polarized photoemission at normal emission^{74–77} or by spin-polarized inverse photoemission at normal incidence,⁷⁸ even for stoichiometric surfaces. Similarly, spin minority surface states leading to a loss of half-metallic character, as noted above, would not generally be

observed if measurements are only undertaken at the $\bar{\Gamma} k_{\parallel} = 0$ point.

Polarization at the Brillouin zone center ($\mathbf{k}_{\parallel} = 0$) corresponds to long range magnetic order but is only loosely related to the magnetic moment. Figure 3 illustrates this point for the polarization of the NiMnSb(100) surface. The data suggest a very high spin polarization near the Fermi level, nearly 100% above background for $\mathbf{k}_{\parallel} = 0$ and $T = 300$ K (Refs. 54 and 73) in spin-polarized inverse photoemission, but as the temperature is increased above 80 K, the MCD indicates a huge decrease in the relative band structure wave vector “averaged” polarization. Angle-resolved spin-polarized photoemission or spin-polarized inverse photoemission measurements, with limited wave vector sampling, cannot be taken as sufficient evidence of half-metallic ferromagnetic character if the sample is crystalline or is polycrystalline with texture growth.

The negligible DOS at the Fermi level [$D(E_F) \sim 0$] in the spin-polarized photoemission spectra of $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$ (Ref. 74) and CrO_2 (Ref. 75) and in spin-polarized inverse photoemission for $\text{La}_{2/3}\text{Pb}_{1/3}\text{MnO}_3$ (Ref. 71) suggests that either the surface is not the correct stoichiometry in those studies (see Ref. 70 for $\text{La}_{2/3}\text{Sr}_{1/3}\text{MnO}_3$, Ref. 72 for $\text{La}_{2/3}\text{Pb}_{1/3}\text{MnO}_3$, Ref. 68 for NiMnSb, and Refs. 94 and 95 for CrO_2) or that the Fermi level crossings are well away from $\bar{\Gamma}$. To a very *small* extent, final state effects in photoemission can also contribute.⁹⁶ A band structure like that of gadolinium, with a strong magnetic surface state near the Fermi energy at $\bar{\Gamma}$, might appear to be a very high polarization ferromagnet, if such measurements are taken at only one wave vector near the surface Brillouin zone center,^{97–99} well below the Debye temperature.⁹⁹ No one would argue that gadolinium is a half-metallic system. Possible crystalline disorder that might occur cannot be used as an argument for “complete” sampling of the Brillouin zone. In such strongly textured thin films, disorder would result in some cylindrical averaging of k points away from $\bar{\Gamma}$ but leave the $\bar{\Gamma}$ point unperturbed. So “proof” of half-metallic character must necessarily remain elusive.

VI. CONCLUSIONS

We have, hopefully, convinced the reader that 98% spin polarization is not 100% spin polarization, even at 1.6 K. Magnons are difficult to suppress, in spite of a fond desire for 100% spin polarization at room temperature. We have shown that finite-temperature spin disorder destroys the complete spin polarization of half-metallic ferromagnets. The many-sublattice structure of the half-metallic ferromagnets, which is most pronounced in zero magnetic field conditions, yields transverse optical phonon modes which couple to thermally excited magnons. In a strict sense, this thermally activated spin mixing means that half-metallic ferromagnetism is limited to perfect crystals at zero temperature. This mechanism is modified and, in general, enhanced by crystal imperfections, and by surface and interface effects.

We do not equate the transport properties of half metals considered in this paper with those observed in ordinary itinerant ferromagnets. In the density of states, the difference is

epitomized by Fig. 1, and the transport is influenced by the contrast between itinerant majority and largely localized minority electrons. In our opinion, half-metallic ferromagnetism is an idealized limit, realistic only in perfect crystals at zero temperature. Due to finite-temperature and surface effects, the elemental ferromagnets (Co, Fe, and Ni) may, in fact, be better for spin injection than half-metallic systems.

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