Theory of record thermopower near a finite temperature magnetic phase transition: IrMn

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The effect of scattering of conduction electrons by dynamical spin fluctuations on the thermopower in metals near a thermal phase transition into an antiferromagnetic phase is considered. We are interested in a transition at room temperature, as has been studied in a heterostructure involving layers of IrMn. We show that the electrical resistivity exhibits a narrow but low peak at the transition, which may be difficult to detect on top of the main contributions induced by phonons and impurities. By contrast, the thermopower is found to exhibit a prominent peak both as a function of temperature $T$ for fixed layer thickness $t_{AFM}$ and as a function of $t_{AFM}$ for fixed $T$. We conjecture that the transition temperature $T_c$ is a function of both $t_{AFM}$ and the Fermi energy $\epsilon_F$. Both dependencies give rise to a sharp peak of the thermopower as a function of $T$ or $t_{AFM}$ near the transition. The estimated magnitude of the peak for the case of three-dimensional longitudinal spin fluctuations is in good agreement with experiment.

I. INTRODUCTION

The recently observed fluctuating temperature dependence of the thermopower in a magnetic heterostructure involving the antiferromagnetic metal IrMn$^1$ suggests that the spin fluctuations near the antiferromagnetic transition may be responsible for the observed peak at the ordering temperature. This finding is all the more interesting in that the magnetic transition temperature may be tuned by the thickness of the IrMn layers to be at room temperature, which makes the effect highly promising for applications. The effect of elastic scattering of conduction electrons by the local spins of magnetic metals near the phase transition into a magnetically ordered phase has been studied first by DeGennes and Friedel$^2$, who assumed the spin configuration to be temperature dependent and static as given by an Ornstein-Zernike form. Somewhat later, Fisher and Langer$^3$ revisited the problem in the light of the theory of classical critical phenomena. These authors pointed out that the equal time spin correlation function entering the scattering cross section also appears in the internal magnetic energy and is therefore related to the specific heat. They also mentioned that inelastic scattering may be important. The effect of inelastic scattering on the thermopower was considered by Entin-Wohlman, Deutscher, and Orbach$^4$ in a model calculation leaving the dynamics of the local spin system as an input quantity to be determined from case to case. A detailed model calculation of the electrical resistivity of antiferromagnetic metals, in the framework of the Self-Consistent Renormalization Theory of spin fluctuations in itinerant magnets$^5, 6$ has been worked out by Ueda$^7$. In all these previous studies the anomalies near the transition found for the transport properties appeared to be relatively weak and cannot account for the prominent peak found in the thermopower as a function of temperature or as a function of layer thickness of an IrMn heterostructure$^1$.

In this paper we estimate the contribution of scattering of the charge carriers by dynamical spin fluctuations by assuming a phenomenological form of the spin excitation spectrum dictated by symmetry and conservation laws. We observe that the presence of gapless fermionic excitations in a metal changes the spin excitation spectrum of the local spins in a decisive way, leading to a strongly temperature dependent, at $T_c$ seemingly divergent contribution to the resistivity. The divergent behavior arises for not too high transition temperature, $T_c \ll \epsilon_F$, where $\epsilon_F$ is the Fermi energy (here and in the following we use energy units of Kelvin). The divergence is cut off close to $T_c$ at a temperature $T_\chi$, marking the transition into a quantum critical regime. The effect may be characterized as a continuation of quantum critical scattering in a narrow region of the phase diagram along the phase boundary. There are two effects introduced by the itinerant electrons into the spin dynamics of antiferromagnetic metals as contrasted to insulators. First, the transition temperature is shifted by an amount proportional to the static wave vector dependent conduction electron spin susceptibility at the ordering wave vector $Q$, $\chi_s(Q,0)$. The latter depends on the Fermi energy and thus gives rise to a dominant contribution to the thermopower. Secondly, the dynamics of the spin fluctuations at low energy is dominated by the Landau damping mechanism leading to inelastic scattering processes strongly enhanced near the transition.

II. MODEL AND METHOD

A. Hamiltonian

We assume a system of interacting localized spins, coupled to conduction electrons as expressed by the Hamil-
tonian

\[ H = H_c + H_S + H_{ex} \]  

(1)

where \( H_c \) represents a single conduction band

\[ H_c = \sum_{k,\alpha} \epsilon_k c_{k\alpha}^\dagger c_{k\alpha}. \]  

(2)

We assume the conduction electron system to be three-dimensional.

The dynamics of the localized spin system is defined by

\[ H_S = \sum_q \sum_{\alpha=x,y,z} I_{\alpha} S_q^{\alpha} S_{-q}^{\alpha}. \]  

(3)

where we allow for anisotropic interaction \( I_z \gg I_{x,y} \)
caused by the strong spin-orbit interaction at the Ir ions.

The coupling of conduction electrons to localized spins is described by

\[ H_{c-S} = J_0 \sum_q s_q S_{-q}. \]  

(4)

Here \( S_q^{\alpha} \) are the Fourier components of the localized spin operators, and the conduction electron spin operator is defined by

\[ s_q = \sum_{k,\alpha} \tau_{\alpha\beta} c_{k+q\alpha} c_{k\beta}, \] 

with \( \tau_{\alpha\beta} \) the vector of Pauli matrices (the coupling constants \( I_{\alpha}, J \) are given in units of [energy/density]).

### B. Spin fluctuations in the paramagnetic phase

The transport properties of the conduction electron system of MnIr in the temperature range around room temperature are governed by electron-phonon interaction and the exchange interaction mediated by \( H_{c-S} \) as we argue below. We assume the system to be anisotropic in spin space, with preferred direction along the \( z \)-axis. We will consider both three-dimensional and two-dimensional spin fluctuations, where the \( 3d \) model appears to describe the experiment [1] better, as we shall see.

In the absence of coupling of local spins and conduction electron spins, for \( J = 0 \), the longitudinal susceptibility of localized spins \( \chi_{loc}^{zz} \) is assumed to be well approximated by the static Ornstein-Zernike form

\[ \chi_{loc}^{zz}(0)(q,\omega) \approx \frac{n_S}{T_z} \frac{1}{(T - T_{c,l})/T_{c,l} + (q - Q)^2 \xi_0^2} \]  

(5)

where \( Q \) is the ordering wave vector and \( \xi_0 \) is a microscopic spin interaction length of the order of a lattice spacing and \( n_S \) is the density of localized spins. The transition to the antiferromagnetic phase is signaled by the divergence of \( \chi_{loc}^{zz}(0) \) at wave vector \( q = Q \) and at temperature \( T_{c,l} = O(1) \). In the quasi two-dimensional slab geometry of IrMn within the nanostructure studied in [1] the transition temperature is found to depend on the layer thickness \( t_{AFM} \). One source of such a dependence is the quantization of the momentum component \( (q - Q)_z \) normal to the layer surface, \( (q - Q)_z = \frac{\pi}{t_{AFM}} (2n + 1) \), \( n = 0, \pm 1, \ldots \). The minimal value of \( (q - Q)_z \) at \( n = 0 \) gives rise to a suppression of the transition temperature

\[ T_{c,l} = T_{c,l} [1 - (\frac{\pi \xi_0}{t_{AFM}})^2] \]  

(6)

In Fig. 1 we show that the \( T_c \) data of Fig. 4 of [1] may be fitted reasonably well by Eq. (6) using a spin interaction length \( \xi_0 = 0.17nm \) and \( t_{AFM} = 308K \). Here the reference transition temperature \( T_{c,l} \) is not the bulk value, which is much higher, but a reduced value appropriate for the composite layer structure. For example, the effective spin exchange energy (the quantity \( I_z \) in our notation) has been found to be strongly varying with the Fermi energy [8], possibly accounting for a reduction of the bulk \( T_c \) value by a factor of \( \sim 3 \). The above estimate differs somewhat from the theoretical results for \( T_c \) versus \( t_{AFM} \) given in [9] and the model calculation for the suppression of \( T_c \) based on the reduction of the interaction volume of a spin near the surface [10].

The conduction electron spin susceptibility in the model of noninteracting fermions is given at low energy by

\[ \chi_s(q,\omega) \approx \chi_s(q,0)(1 + i\pi \frac{\omega}{\nu_p q}). \]  

(7)
where \( v_F \) is the Fermi velocity (we employ an isotropic band structure for simplicity) and we assume isotropy in spin space. In the presence of coupling the susceptibility of localized spins is renormalized approximately as

\[
\chi_{loc}^{zz}(q, \omega) \approx \frac{1}{[\chi_{loc}^{zz}(0)(q, \omega)]^{-1} + J^2 \chi_z(q, \omega)} (8)
\]

In the vicinity of the transition and for small \( \omega \) the susceptibility \( \chi_{loc}^{zz} \) is sharply peaked at \( q \approx Q \) so that \( \chi_z(q, \omega) \) may be replaced by \( \chi_z(Q, \omega) \), and \( \chi_{loc}^{zz} \) takes the form

\[
\chi_{loc}^{zz}(q, \omega) \approx \frac{n_s}{T_z} \frac{1}{\tau + (q - Q)^2 \xi_0^2 + \frac{\omega}{\gamma}} (9)
\]

Here we defined the reduced temperature \( \tau = (T/T_c - 1) \) and a renormalized transition temperature \( T_c = T_{c,1} - \Delta T_c \) where \( \Delta T_c = T_{c,1} n_S J^2 \chi_z(Q,0)/I_z \approx T_{c,f}(n_S J^2/I_z) N_F \) with \( N_F = mk_F/\pi^2 \) the conduction electron density of states at the Fermi level (\( m \) is the mass and \( k_F \) is the Fermi wave number of the conduction electrons). The transition temperature \( T_c \) is seen to depend on the Fermi energy \( v_F = k_F^2/2m \), a fact of considerable importance for the thermopower as we show later. The reference energy \( \gamma \) entering the Landau damping term in the denominator of \( \chi_{loc}^{zz}(q, \omega) \) is defined as \( \gamma = [(n_S J^2/I_z) \chi_z(Q,0)]^{-1} v_F Q/\pi \approx (I_z/\pi n_S J^2 N_F) v_F Q \approx T_c (N_F J)^{-2} \), assuming \( Q \approx k_F \) and \( n_s \approx n_s \approx N_F \).

C. Spin fluctuations in the antiferromagnetic phase

In the ordered phase the translation invariance is broken as indicated by the ordering vector \( Q \). The rotation invariance in spin space is also broken by the appearance of the ordered moment vector \( M(R_f) \).

Spin fluctuations along \( M \) (longitudinal) and orthogonal to \( M \) (transverse) behave in a fundamentally different way. The magnetic order also affects the quasiparticle excitations in major ways. The Brillouin zone gets shrunk into the magnetic Brillouin zone, leading to multiple bands induced by back-folding. To keep things simple we will assume two-sublattice antiferromagnetic order \( (Q = (\pi, \pi, \pi)) \) for which case one finds two bands, dubbed "valence" and "conduction" band, separated by a gap \( \Delta \). The structure of the quasiparticle and collective excitations has been calculated for the Hubbard model within the Random Phase Approximation [11][13].

1. Longitudinal spin fluctuations

The longitudinal spin susceptibility is finite in the ordered phase but diverges upon approach to the thermal transition. This is expressed by the form

\[
\chi_{loc}^{zz}(q, \omega) \approx \frac{n_s}{T_z} \frac{1}{\tau_I (q - Q)^2 \xi_0^2 + i(\omega/\gamma)}, (10)
\]

where \( r_I = c_l M_s^2 = c_l M_0^2 |\tau| \), assuming mean field behavior of the ordered moments \( M_s \propto M_0 |\tau|^{1/2} \), where \( M_0 \) is the saturated moment. Here \( c_l \) is a constant of \( O(1) \).

2. Transverse spin fluctuations

If spin rotation invariance in the plane orthogonal to the ordered moments \( M(R_f) \) is still preserved, which we suspect not to be the case for MnIr, gapless spin excitations (spin waves) exist. In this case the transverse spin susceptibility involving spin excitations orthogonal to \( M(R_f) \) is divergent in the whole ordered phase, not just at the critical point. More generally, the transverse spin susceptibility may be expressed as

\[
\chi^{zz}(q, \omega) \approx \frac{n_s}{T_z} \frac{1}{\Delta_{zz} + (q - Q)^2 + i(\omega/\gamma)} (11)
\]

where \( \Delta_{zz} \) is the spin wave gap. The coupling of the transverse spin fluctuations to the quasiparticles depends on the respective quasiparticle bands. For definiteness we assume that the Fermi energy lies in the lower (valence) band. Then the scattering of quasiparticles from the valence band into the valence band is suppressed, the corresponding vertex \( A_{uu}^{zz}(k, k + q + Q) \propto q \cdot \nabla \epsilon_k \) such that the product \( (A_{uu}^{zz})^2 \chi^{zz} \) appearing in the expression for the self energy is no longer singular and hence does not give rise to critical behavior [12][14]. By contrast, the vertex function for scattering from the valence band into the conduction band is of \( O(1) \). However, the latter excitation requires a minimum energy \( \Delta \) and is thus possible only in the vicinity of the transition when \( T \gtrsim \Delta \). In the following we will assume that the spin wave gap \( \Delta > T_c \) that spin waves may not be thermally excited.

III. ELECTRICAL RESISTIVITY

We now present a model calculation of the contribution of scattering by antiferromagnetic fluctuations to the electrical resistivity \( \rho \). Since the typical momentum transfer in such scattering processes is large, of order of the ordering wave vector \( Q \), and therefore, assuming a half-filled conduction band, of order \( k_F \), the momentum relaxation rate \( 1/\tau_{tr} \) is approximately related to the imaginary part of the electron self energy \( \Sigma(k, \omega) \) by \( 1/\tau_{tr} \approx 2Im \Sigma \). The resistivity is then given in terms of the self energy as

\[
\rho \approx \frac{m}{e^2 n} 2Im \Sigma(k_F, \omega \approx T) (12)
\]
A. Three-dimensional spin fluctuations

In a clean metal at low temperature scattering by antiferromagnetic fluctuations affects only a small part of the Fermi surface: the "hot spots" connected by the ordering wave vector $\pm Q$. At higher temperatures phonon scattering, or at all temperatures impurity scattering, helps to remove the constraints imposed by momentum conservation so that the critical behavior at the hot spots is distributed all over the Fermi surface. The self-energy may be calculated to one-loop order, taking into account phonon and impurity scattering, which leads to a prefactor $A = O(1)$.

In the case of electrons and spin fluctuations that are both three-dimensional, we get

$$\text{Im} \Sigma(k, \omega) \approx A J^2 \int_{-\infty}^{\infty} \frac{d\nu}{2\pi} \int \frac{d^3q}{8\pi^3} \text{Im} G(k + q + Q, \omega + \nu) \times \text{Im} \chi(q + Q, \nu) |f(\nu + \omega) + b(\nu)|$$  \hspace{1cm} (13)

where $f(\omega), b(\nu)$ are the Fermi and Bose functions respectively, and $J$ is the coupling constant of electrons and spin fluctuations. The conduction electron spectral function is approximated by

$$\text{Im} G(k, \omega) = \pi \delta(\omega - \epsilon_k)$$  \hspace{1cm} (14)

We will employ an isotropic model of the conduction band with energy $\epsilon_k = k^2/2m - \epsilon_F$, for which the angular integral may be done, assuming $q \ll Q$, with the result

$$\frac{1}{2} \int_{-1}^{1} d\cos \theta \text{Im} G(k + q + Q, \omega + \nu) \approx \frac{\pi}{2} \frac{m}{k_F Q} \approx \frac{1}{\epsilon_F}$$  \hspace{1cm} (15)

At low frequency $\omega < T$ we may drop $\omega$ in the argument of the Fermi function, whence $\text{Im} \Sigma(k, \omega)$ is approximately independent of frequency

$$\text{Im} \Sigma(k, \omega \ll T) \approx A \frac{n s J^2}{\epsilon_F} \int_{-\infty}^{\infty} \frac{d\nu}{2\pi \sinh(\nu/T)} \times \int_{0}^{\infty} \frac{dq q^2}{2\pi^2} \frac{\nu/\gamma}{(\nu + (q/k_F)^2)^2 + (\nu/\gamma)^2}$$  \hspace{1cm} (16)

The divergence of the momentum integral at $q \to 0$ is cut off in two ways: (i) in the limit $T \to 0$ the Landau damping term provides the cutoff just as is the case at the quantum critical point (QCP). The ensuing quantum critical behavior is therefore found to extend in the phase diagram from the QCP along the phase boundary in a narrow strip of width $\tau_x T_c$, where $\tau_x \approx T_c/\gamma$; (ii) in the limit $\nu \to 0$, or more generally $T \to 0$ (considering that the frequency integral is confined to $|\nu| \lesssim T$) the cutoff is provided by the term $\tau$. This is the quantum disordered regime, in which $\text{Im} \Sigma$ is found to depend critically on $\tau$. The latter behavior may be seen in analogy to the approach to the QCP at $T = 0$ from the quantum disordered side.

Provided the transition temperature is not too high, $T_c \ll \gamma$, there exists a wide regime of reduced temperatures $\tau_x \ll \tau \ll 1$ with $\tau_x \approx T_c/\gamma$ for which $\text{Im} \Sigma$ is approximately given by

$$\text{Im} \Sigma(k, \omega \ll T) \approx \frac{3}{2\pi} \frac{n s J^2 T^2}{\epsilon_F \tau_x} \int_{0}^{T} d\nu \int_{\sqrt{\nu}}^{\infty} \frac{dq/k_F^2}{(q/k_F^2)^2 + (\nu/\gamma)^2}$$

$$\approx \frac{3}{2\pi} \frac{n s J^2 T^2}{\epsilon_F \tau_x} \frac{1}{\epsilon_F \gamma \sqrt{\nu}}$$  \hspace{1cm} (17)

where $n = k_B^2/3\pi^2$ is the density of conduction electrons. The resistivity follows as

$$\rho_{sfl}^{3d} = \rho_0 \frac{1}{\sqrt{\tau_x}}, \quad \tau_x \ll \tau$$  \hspace{1cm} (18)

where

$$\rho_0 \approx \frac{9}{4} R_Q k_F^2 \frac{n s J n J}{T_x} \frac{T_x^2}{\epsilon_F \gamma},$$

$$R_Q = \frac{h}{e^2} \approx 2581 \Omega$$

and $R_Q = \frac{h}{e^2} \approx 2581 \Omega$ is the quantum resistance. In the ordered phase, in the temperature regime $\tau_x \ll |\tau| < 1$, where $\tau_x, l \approx T_c/\gamma$, with $\gamma = \gamma_c M_0^2$, the resistivity scale $\rho_0$ in Eq. (18) is replaced by $\rho_0 = \rho_0/(M_0 \sqrt{\gamma})$. In the temperature regime close to the transition, defined by $-\tau_x < \tau < \tau_x$, the resistivity is given by

$$\rho_{sfl}^{3d} = \rho_0 \frac{1}{\sqrt{\tau_x}} \approx -\tau_x < \tau < \tau_x$$  \hspace{1cm} (19)

We now estimate the $\rho_{sfl}^{3d}$ of IrMn layers as studied in [1]. The reference resistance setting the scale is given by $R_Q k_F^2 \approx 258 \Omega \sqrt{\mu}$ cm taking $k_F \approx 10^3 \text{cm}^{-1}$. The remaining factors at the transition temperature are $\gamma \approx 10^4 K$, $T_c/\gamma \approx 0.03$, $\tau_x \approx T_c \approx 300 K$, $n J \approx N_F J \epsilon_F \approx 1.5 \times 10^3 K$, assuming $\epsilon_F \approx 10^4 K$, and $A \approx 1$, resulting in $\rho_0 \approx 0.4 \Omega \text{cm}$. This is very small in comparison to the observed resistivity (Fig.2 of the Supplementary information of Tu et al. [1] where $\rho \approx 170 \Omega \text{cm}$ at $T_c$). Indeed, the resistivity data do not show any trace of a peak at $T_c$. Here we anticipate that the crossover scale $\tau_x \approx 0.06$ (see below), leading to the estimate of the maximum height of the spin fluctuation induced contribution as $\rho_{sfl}(T_c) = \rho_0 \frac{1}{\sqrt{\tau_x}} \approx 1.5 \Omega \text{cm}$. The model calculations for bulk MnIr [2] show a hump in the electrical resistivity below $T_c$.

B. Two-dimensional spin fluctuations

In this case the self-energy at low frequency is found as
Using $n = k_F^2/3\pi^2$. We note that magnetic order is not destroyed by transverse spin fluctuations because those are gapped out. The resistivity shows a more strongly divergent behavior for $\tau \to 0$, up to the crossover temperature $\tau_x$

$$\rho_{sfl}^{2d} = \rho_0 \frac{\pi}{k_Ft_{AFM}} \tau \left\{ \begin{array}{l} 1, \quad \tau \gg \frac{\tau_x}{\tau_{c,l}}, \\
1/\xi_0, \quad \tau \gg \tau_x, \end{array} \right. \quad (23)$$

The maximum of $\rho_{sfl}^{2d}$ is reached for $\tau < \tau_x$ and may be estimated as $\rho_{sfl}^{2d} \approx \rho_0 \frac{\pi}{k_Ft_{AFM}} \frac{1}{\tau_x} \approx 0.5 \mu\Omega cm$, assuming $t_{AFM} = 30 \AA$, which is again much less than the total resistivity.

The system showing a record thermopower studied in [1] consists of layers of IrMn of thickness varying between 0.6 - 4 nm and is therefore quasi two-dimensional. The momentum component $q_z$ of the spin fluctuations normal to the sample plane is quantized, $q_z = n(2n + 1)$, $n = 0, 1, 2,...$, where $t_{AFM}$ is the sample thickness. For sufficiently small thickness $t_{AFM}$ only the lowest transverse mode is occupied. For this to be valid we should have $(2\pi t_{AFM} \xi_0)^2 \gg \tau_x$, where $\xi_0 \approx k_F^{-1}$ is the microscopic spin interaction length. For the IrMn layers of thickness $\approx 3 nm$ the latter condition is not satisfied, taking $\xi_0 \approx 0.17 nm$. We therefore conclude that the three-dimensional model is more appropriate for describing the sample with $t_{AFM} = 2.8 nm$, for which a detailed comparison with the thermopower data is possible (see below).

### IV. THERMOPOWER

The thermopower is defined by

$$S = -\frac{2k_B^2 T \rho'(\epsilon_F)}{\epsilon \rho(\epsilon_F)} \quad (24)$$

where $\rho'(\epsilon_F) = d\rho/d\epsilon_F \approx d\rho_{sfl}/d\epsilon_F$, assuming that of all contributions to the resistivity, by scattering off phonons, off impurities, off magnetic ions, the largest contribution is coming from critical spin fluctuations.

The essential dependence of $\rho_{sfl}$ on $\epsilon_F$ is through the shift of the transition temperature $T_c$ induced by changing $\epsilon_F$. Using $d\tau/d\epsilon_F = -(T/T_c^2)dT_c/d\epsilon_F$, one then finds above the transition in case of three- or two-dimensional spin fluctuations

$$\text{Im} \Sigma(k, \omega \lesssim T) \approx \frac{A}{2\pi^2} \frac{n_s J^2}{I_\gamma} \int_{-\infty}^{\infty} \frac{d\nu}{\sinh(\nu/T)} \times \int \frac{dq/t_{AFM}}{(\tau + (q/k_F)^2)^2 + (\nu/\gamma)^2} \quad (21)$$

$$\approx \frac{3}{2} \frac{A}{2} \frac{n_s J^2}{I_z} \frac{T^2}{\gamma \epsilon_F} \frac{1}{\tau_{AFM}} \quad (22)$$

a power law divergence $S \propto (T - T_c)^{-\alpha}$, which is cut off at $\tau \approx T_c/\epsilon_F$. The dependence of $T_c$ on $\epsilon_F$ has been discussed above, $dT_c/d\epsilon_F \approx (n_s J/I_z)(JN_F)/(T_c/\epsilon_F) \approx (N_FJ)^2 \approx 0.02$. The peak height follows, using these estimates and $k_B \epsilon_F \approx 8.6 \times 10^{-3} V/K$, as $S(T_c) \approx 240 \mu V/K$, taking $\tau_x = 0.06$, for both three- or two-dimensional fluctuations, which is of the order of magnitude observed in experiment, at least for the compounds containing only IrMn magnetic layers. The compounds consisting of additional adjacent CoFeB magnetic layers show even stronger thermopower, possibly because the coupling constants $J, I$ are effectively changed by the magnetic environment (increased $J$, decreased $I_z$). For temperatures below the transition the prefactor $\rho_0$ and the cutoff scale $\tau_x$ are replaced by $\rho_{0,l}$ and $\tau_{x,l}$.

In Fig. 2 we show a comparison of our theory with the Seebeck voltage data of a sample with IrMn layer thickness of $t_{AFM} = 2.8 nm$. The transition temperature is chosen to be $T_c = 281 K$, slightly less than the 285K estimated in [1]. We use an interpolation expression connecting the quantum critical regime at $|\tau| < \tau_x$ and the quantum disordered regime at $|\tau| > \tau_x$ at $T > T_c$: $|\tau|\Theta(\tau_x - |\tau|) + \tau_x \Theta(\tau_x - |\tau|) \approx \sqrt{\tau^2 + \tau_x^2}$ (here $\Theta(x)$ is the unit step function). A similar expression is employed for $T < T_c$. We find that the best fit is obtained assuming three-dimensional spin fluctuations ($d = 3$).
Seebeck voltage $V_S$ at $T \gtrsim T_c$ is then described by

$$V_S^{>;<} = c_1^{>;<} \left(\frac{T}{T_c}\right)^{2(\frac{T}{T_c} - 1)^2} + (\frac{T}{T_c} - 1)^2 \right)^{-0.75} + c_2^{>;<},$$

(26)

The parameters used for fitting Eq. (26) to the data of Fig. 2 of [1] are $T^0_c = 209K$, $T_c^0 = 269.5K$, $c_1^0 = 0.89mV$, $c_2^0 = 0.4mV$, $c_2^2 = -6.5mV$, and $c_2^2 = 0mV$. The constants $c_2^{>;<}$ account for contributions derived from other scattering processes (phonons, impurities, magnetic ions) or form $d\rho_0/d\epsilon_F$.

V. CONCLUSION

Usually the spin fluctuations in a spin system near a continuous thermal phase transition into a magnetically ordered state are classical. In a metal, however, the dynamics may have quantum character even at finite temperature on account of coupling of the localized spins to the conduction electron spins, giving rise to Landau damping. Provided the transition temperature is not too low, $T_c \ll \gamma$, where $\gamma$ is the characteristic energy scale of the Landau damping, quantum fluctuations may give rise to quantum critical behavior. As we have shown here, the electrical resistivity acquires a quantum critical contribution following a power law divergence in the reduced temperature $|\tau_\gamma|^\alpha$ up to a crossover scale $\tau_\gamma \ll 1$. While the corresponding peak structure in the resistivity may be small in comparison to the dominant terms due to scattering by phonons, magnetic moments or impurities, it gives rise to a prominent peak in the thermopower. This is because the critical temperature $T_c$ is found to depend sufficiently strongly on the Fermi energy. We compare our theoretical results with recent experimental observations [1] on a heterostructure containing the antiferromagnetic metal IrMn in the form of layers of several nanometers thickness, and find excellent agreement. The strong dependence of the critical temperature on the layer thickness found in [1] may also be explained by our theory.

While we have focussed on the experiments on IrMn, the results suggest that large values of thermopower may result in other metallic films from the coupling to magnetic fluctuations. The current experimental results argue for fluctuations that are still in the three-dimensional regime, both from the form of Figure 1, where the reduction by finite size of $T_{c,I}$ from the limit $T_{c,I}$ is modest compared to the cutoff $\tau_x = T_c/\gamma$ for the films of interest, and from the fit to the thermopower data in Fig. 2. Note that we have argued that the much greater reduction in critical temperature from bulk IrMn is due to a change in the microscopic parameters (e.g. $f_x$) for the layers rather than a finite-size effect, in the language of critical phenomena. One might expect even stronger enhancement at room temperature by effective two-dimensionality, provided films of some material are thin enough that the relative reduction due to finite size, i.e. as in Equation 4 is greater than the cut-off $\tau_x$. For a fixed critical temperature smaller values of $\tau_x$ would require a larger Landau damping parameter $\gamma$, which seems unlikely. The effect of quantum fluctuations at a finite temperature antiferromagnetic transition has also been seen in specific heat data for two heavy fermion metals, CeCu$_{6-x}$Au$_x$ [15] and YbRh$_2$Si$_2$ [16] as will be shown in upcoming work [17].

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[1] S. Tu, T. Ziman, G. Yu, C. Wan, J. Hu, H. Wu, H. Wang, M. Liu, C. Guo, J. Zhang, M. A. Cabero Z., Y. Zhang, P. Gao, S. Liu, D. Yu, X. Han, I. Hallsteinsen, D. A. Gilbert, P. Wölfle, K. L. Wang, J-P. Ansermet, S. Maekawa, and H. Yu, Nature Commun. 11, 2023 (2020).
[2] P. G. De Gennes, and J. Friedel, J. Phys. Chem. Solids 4, 71 (1958).
[3] M. E. Fisher, and J. S. Langer, Phys. Rev. Lett. 20, 665 (1968).
[4] O. Entin-Wohlman, G. Deutscher, and R. Orbach, Phys. Rev. B 14, 4015 (1976).
[5] H. Hasegawa, and T. Moriya, J. Phys. Soc. Jpn. 36, 1542 (1974).
[6] T. Moriya, Spin Fluctuations in Itinerant Electron Magnetism, (Springer, New York, 1985).
[7] K. Ueda, J. Phys. Soc. Jpn. 43, 1497 (1977).
[8] R. Y. Umetsu, M. Miyakawa, K. Fukamichi, and A. Sakuma, Phys. Rev. B 69, 104411 (2004).
[9] L. Frangou, S. Oyarzun, S. Auffret, L. Vila, S. Gamberelli, and V. Baltz, Phys. Rev. Lett. 116, 077203 (2016).
[10] R. Zhang and R. F. Willis, Phys. Rev. Lett. 86, 2665 (2001).
[11] T. Moriya, Spin Fluctuations in Itinerant Electron Magnetism, Springer Series in Solid-State Sciences (Springer Berlin Heidelberg, 2012).
[12] J. R. Schrieffer, X. G. Wen, and S. C. Zhang, Phys. Rev. B 39, 11663 (1989).
[13] A. V. Chubukov and D. M. Frenkel, Phys. Rev. B 46, 11884 (1992).
[14] S. Adler, Phys. Rev. 137 B, 1022, (1965).
[15] H. v. Löhneysen, T. Pietrus, G. Portisch, H. G. Schlager, A. Schröder, M. Sieck, and T. Trappmann, Phys. Rev. Lett. 72, 3262 (1994).
[16] C. Krellner, S. Hartmann, A. Pikul, N. Oeschler, J. G. Donath, C. Geibel, F. Steglich, and J. Wosnitza, Phys. Rev. Lett. 102, 196402 (2009).

[17] P. Wölfle, and J. Schmalian, unpublished.