



## Transport measurements of the spin-wave parameters of thin Mn films

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Temperature-dependent transport measurements on ultrathin antiferromagnetic Mn films reveal a heretofore unknown nonuniversal weak-localization correction to the conductivity which extends to disorder strengths greater than 100 k $\Omega$  per square. The inelastic scattering of electrons off of gapped antiferromagnetic spin waves gives rise to an inelastic scattering length which is short enough to place the system in the three-dimensional regime. The extracted fitting parameters provide estimates of the energy gap ( $\Delta \approx 16$  K) and Heisenberg exchange constant ( $J \approx 1000$  K).

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### I. INTRODUCTION

Thin-film transition metal ferromagnets (Fe, Co, Ni, Gd) and antiferromagnets (Mn, Cr) and their alloys are not only ubiquitous in present day technologies but are also expected to play an important role in future developments [1]. Understanding magnetism in these materials, especially when the films are thin enough so that disorder plays an important role, is complicated by the longstanding controversy about the relative importance of itinerant and local moments [2–4]. For the itinerant transition metal magnets, a related fundamental issue centers on the question of how itinerancy is compromised by disorder. Clearly with sufficient disorder the charge carriers become localized, but questions arise as to what happens to the spins and associated spin waves and whether the outcome depends on the ferro/antiferro alignment of spins in the itinerant parent.

Ferromagnets, which have magnetization as the order parameter, are fundamentally different than antiferromagnets, which have staggered magnetization (i.e., difference between the magnetization on each sublattice) as the order parameter [5]. Ferromagnetism thus distinguishes itself by having soft modes at zero wave number, whereas antiferromagnets have soft modes at finite wave number [6]. Accordingly, the respective spin-wave spectrums are radically different. These distinctions are particularly important when comparing quantum corrections to the conductivity near quantum critical points for ferromagnets [7] and antiferromagnets [8].

Surprisingly, although there have been systematic studies of the effect of disorder on the longitudinal  $\sigma_{xx}$  and transverse  $\sigma_{xy}$  conductivity of ferromagnetic films [9–13], there have been few if any such studies on antiferromagnetic films. In this paper we remedy this situation by presenting transport data on systematically disordered Mn films, finding an unexpected three-dimensional (3D) weak-localization correction to the conductivity extending to disorder strengths greater than 100 k $\Omega$ . In contrast, the localization corrections are always in two dimensions (2D) for thin ferromagnetic films.

### II. EXPERIMENTAL METHODS AND BACKGROUND

Our Mn films with square shapes 0.9 mm on a side are sputter deposited through a Hall cross shadow mask onto

polished sapphire substrates in a custom-designed vacuum chamber and then transferred without exposure to air into an adjacent cryostat for transport studies to low temperature. The experimental procedures are similar to those reported previously: disorder, characterized by the sheet resistance  $R_0$  measured at  $T = 5$  K using dc techniques, can be changed either by growing separate samples or by gentle annealing of a given sample through incremental stages of disorder [14].

In this study we focus on a single *in situ* prepared Mn film with initial disorder strength  $R_0 \approx 6$  k $\Omega$ . The temperature dependence of the resistance was measured in the temperature range 5–50 K. Upon heating the sample for short periods to above 150 K, we observed irreversible incremental increases of resistance which remained stable for temperatures below 50–100 K. Higher disorder strengths required higher temperature anneals. This annealing procedure thus allowed us to increase the disorder strength  $R_0$  for a single film in a controllable way and in small steps. Subsequent to each anneal, the temperature dependence of the resistance was again measured in the temperature range 5–50 K.

We suspect that at each annealing temperature there is a transfer of a small amount of material from the thinner to the thicker portions of the film. In the limit where a film can be considered as granular, annealing would cause individual grains to incrementally pucker up and increase their height while simultaneously increasing their separation, thereby increasing the intergranular resistance. In addition, quench cooling from the higher temperature anneals might freeze in higher degrees of disorder. To check the results for reproducibility, we prepared two additional films with intermediate disorder (about 10 and 20 k $\Omega$ ). These films, measured as grown without annealing, gave the same temperature dependence as the annealed sample. Similar checks were performed in our earlier work on ferromagnetic Gd films, where we found that films with a given sheet resistance  $R_0$ , obtained either by annealing or by a single deposition, had very similar temperature dependencies [14]. Accordingly,  $R_0$  provides a robust and reproducible characterization of disorder.

Using these same experimental procedures as described for ferromagnetic Gd films [14], our results for antiferromagnetic Mn, however, are decidedly different. The data are well described over a large range of disorder strengths by a nonuniversal 3D quantum correction that applies only to spin-wave gapped antiferromagnets. This finding implies the presence

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of strong inelastic electron scattering off of antiferromagnetic spin waves. The theory is validated not only by good fits to the data, but also by extraction from the fitting parameters of values for the spin-wave gap  $\Delta$  and the exchange constant  $\bar{J}$  that are in agreement with the values expected for Mn.

In previous work the inelastic scattering of electrons off of spin waves has been an essential ingredient in understanding disordered ferromagnets. For example, to explain the occurrence of weak-localization corrections to the anomalous Hall effect in polycrystalline Fe films [11], it was necessary to invoke a contribution to the inelastic phase breaking rate  $\tau_\varphi^{-1}$  due to spin-conserving inelastic scattering off spin-wave excitations. This phase breaking rate, anticipated by theory [15] and seen experimentally in spin-polarized electron energy loss spectroscopy (SPEELS) measurements of ultrathin Fe films [16,17], is linear in temperature and significantly larger than the phase breaking rate due to electron-electron interactions, thus allowing a wide temperature range to observe weak-localization corrections [11]. The effect of a high  $\tau_\varphi^{-1}$  due to inelastic scattering off spin-wave excitations is also seen in Gd films, where in addition to a localizing  $\log(T)$  quantum correction to the conductance, a localizing linear-in- $T$  quantum correction is present and is interpreted as a spin-wave-mediated Altshuler-Aronov type correction to the conductivity [12].

Interestingly, this high rate of inelastic spin-rate scattering becomes even more important for the thinnest films, as shown in theoretical calculations on Fe and Ni which point to extremely short spin-dependent inelastic mean free paths [18] and in SPEELS measurements on a-few-monolayers-thick Fe/W(110) films in which a strong nonmonotonic enhancement of localized spin-wave energies is found on the thinnest films [17].

Inelastic spin-wave scattering in highly disordered ferromagnetic films can be strong enough to assure that the associated  $T$ -dependent dephasing length  $L_\varphi(T) = \sqrt{D\tau_\varphi}$  (with  $D$  the diffusion constant) [19] is less than the film thickness  $t$ , thus putting thin films into the 3D limit where a metal-insulator transition is observed [14]. Recognizing that similarly high inelastic scattering rates must apply to highly disordered antiferromagnetic films, we first proceed with a theoretical approach that takes into account the scattering of antiferromagnetic spin waves on the phase relaxation rate and find a heretofore unrecognized nonuniversal 3D weak-localization correction to the conductivity that allows an interpretation of our experimental results.

We mention in passing that the 3D interaction-induced quantum correction found to be dominant in the case of *strongly disordered* ferromagnetic Gd films, which undergo a metal-insulator transition [14], is found to be much smaller in the present case and will not be considered further. (For an estimate of this contribution, see Ref. [20].)

### III. THEORY—WEAK LOCALIZATION IN 3D ANTIFERROMAGNETS

As discussed in detail in Ref. [21], the phase relaxation time  $\tau_\varphi$  limits the phase coherence in a particle-particle diffusion

propagator  $C(q, \omega)$  (Cooperon) in the form

$$C(q, \omega_n) = \frac{1}{2\pi N_0 \tau^2} \frac{1}{Dq^2 + |\omega_n| + 1/\tau_\varphi}, \quad (1)$$

where  $N_0$  is the density of states at the Fermi level,  $\tau$  is the elastic scattering time, and  $\omega_n = 2\pi nT$  is the Matsubara frequency. Labeling the Cooperon propagator in the absence of interactions as  $C_0$ , we can write

$$\frac{1}{\tau_\varphi} = \frac{1}{2\pi N_0 \tau^2} [C^{-1} - C_0^{-1}]. \quad (2)$$

In general,  $C(q, \omega)$  can be evaluated diagrammatically in the presence of interactions and disorder in a ladder approximation [22] that can be symbolically written as  $C = C_0 + C_0 K C$ , where the interaction vertex  $K$  contains self-energy as well as vertex corrections due to both interactions and disorder. It then follows that  $1/\tau_\varphi$  is given by

$$\frac{1}{\tau_\varphi} = -\frac{1}{2\pi N_0 \tau^2} K. \quad (3)$$

In Ref. [21], the leading temperature and disorder dependence of the inelastic diffusion propagator was evaluated diagrammatically, in the presence of ferromagnetic spin-wave-mediated electron-electron interactions. Here we consider the antiferromagnetic case. We only consider the large spin-wave gap where the damping can be ignored. Using the antiferromagnetic dispersion relation  $\omega_q = \Delta + A_s q$ , where  $A_s$  is the spin stiffness, the inelastic lifetime is given by

$$\frac{\hbar}{\tau_\varphi} = \frac{4\bar{J}^2}{\pi \hbar n} \int_0^{1/l} \frac{q^{d-1} dq}{\sinh \beta \omega_q} \frac{Dq^2 + 1/\tau_\varphi}{(Dq^2 + 1/\tau_\varphi)^2 + \omega_q^2}, \quad (4)$$

where  $n = k_F^3/3\pi^2$  is the 3D carrier density,  $\bar{J}$  is an exchange constant, and  $\beta = 1/k_B T$ . Here we will consider the limit  $\hbar/\tau_\varphi \ll \Delta$ , relevant for our experiment on Mn. In this limit we can neglect the  $1/\tau_\varphi$  terms inside the integral. The upper limit should be restricted to  $\Delta/A_s$  in the limit  $\Delta/A_s < 1/l$ , where  $l$  is the elastic mean free path. For large disorder, we expect the parameter  $x \equiv \hbar D k_F^2 \Delta / \bar{J}^2 \ll 1$ , where the spin-exchange energy is related to the spin stiffness by  $\bar{J} = A_s k_F$ . In this limit,  $L_\varphi$  can be simplified as

$$k_F L_\varphi \approx \left(\frac{\bar{J}}{\Delta}\right)^{3/2} \left(\frac{5 \sinh \frac{\Delta}{T}}{12\pi}\right)^{1/2}, \quad x \ll 1, \quad (5)$$

which is independent of  $x$ , and therefore, independent of disorder.

Given the inelastic lifetime, the weak-localization correction in 3D is usually given by [19]  $\delta\sigma_{3d} = e^2/\hbar\pi^3 L_\varphi$ , where the prefactor to the inverse inelastic length is a universal number, independent of disorder. However, at large enough disorder, we show that there exists a disorder-dependent correction, due to the scale-dependent diffusion coefficient near the Anderson metal-insulator transition. In fact, the diffusion coefficient obeys the self-consistent equation [23]

$$\frac{D_0}{D(\omega)} = 1 + \frac{k_F^{2-d}}{\pi m} \int_0^{1/l} dQ \frac{Q^{d-1}}{-i\omega + D(\omega)Q^2}, \quad (6)$$

where  $D_0 = v_F l/d$  is the diffusion coefficient at weak disorder. While the significance of the prefactor to the integral

is not clear, the above equation remains qualitatively accurate over a wide range near the Anderson transition. Setting  $\omega = i/\tau_\varphi$  and doing the  $Q$  integral in 3D,

$$\begin{aligned} \frac{D_0}{D} &\approx 1 + \frac{1}{\pi m k_F} \int_{1/L_\phi}^{1/l} dQ \frac{Q^2}{D Q^2} \\ &= 1 + \frac{D_0}{D} \frac{3}{\pi k_F^2 l^2} - \delta \left( \frac{D_0}{D} \right), \end{aligned} \quad (7)$$

where

$$\delta \equiv \frac{D_0}{D} \frac{3}{\pi k_F^2 l^2} \frac{l}{L_\phi} \quad (8)$$

is assumed to be a small correction, and Eq. (7) should not be solved self-consistently. This follows from the fact that the diffusion coefficient of electrons at fixed energy entering the Cooperon expression is that of noninteracting electrons and is given by the limit  $T \rightarrow 0$ ,  $L_\phi \rightarrow \infty$  and therefore  $\delta \rightarrow 0$ . Then the correction at finite  $T$  is given by

$$\begin{aligned} \frac{D}{D_0} &= \frac{1}{\left(\frac{D_0}{D}\right)_0 - \delta \left(\frac{D_0}{D}\right)} \\ &\approx \left(\frac{D}{D_0}\right)_0 + \left(\frac{D}{D_0}\right)_0 \frac{3}{\pi k_F^2 l^2} \frac{l}{L_\phi}, \end{aligned} \quad (9)$$

where

$$\lim_{T \rightarrow 0} \frac{D}{D_0} \equiv \left(\frac{D}{D_0}\right)_0. \quad (10)$$

Using the relation  $\sigma_{3d} = e^2 N_0 D$  where the longitudinal sheet conductance  $\sigma_\square = \sigma_{3d} t$ , with  $t$  being the film thickness, we finally get the temperature-dependent weak-localization correction term

$$\begin{aligned} \frac{\delta \sigma_\square}{L_{00}} &= \left(\frac{D}{D_0}\right)_0 \frac{2}{\pi} \frac{t}{L_\phi} \\ \left(\frac{D}{D_0}\right)_0 &\approx \frac{2}{1 + \sqrt{1 + 4R_0^2/a^2}}, \end{aligned} \quad (11)$$

where  $R_0 = L_{00}/\sigma_\square(T=0)$ ,  $L_{00} = e^2/\pi h$ ,  $a = 3\pi/2k_F t b_0$ ,  $b_0$  is a number of order unity, and we have solved the self-consistent equation for  $D$  in order to express  $D_0$  in terms of  $D$  and finally  $R_0$ . Thus in this case, the weak-localization correction has a prefactor which is not universal. While this reduces to the well-known universal result at weak disorder  $R_0 \ll a$ , it becomes dependent on disorder characterized by the sheet resistance  $R_0$  at strong disorder and at the same time substantially extends the 3D regime near the transition. A weak-localization correction at strong disorder has not been identified before. Here it is observable due to the extremely small thickness of the films, boosting the parameter  $a$ . For thicker films the weak-localization correction is unobservably small, except at weak disorder.

#### IV. EXPERIMENTAL RESULTS AND DISCUSSION

Using the expression for  $L_\varphi$  [Eq. (5)] into Eq. (11), we finally obtain the total conductivity, including the quantum correction to the conductivity due to weak localization in 3D

arising from scattering of electrons off antiferromagnetic spin waves in Mn,

$$\frac{\sigma_\square}{L_{00}} = A + \frac{B}{\sqrt{\sinh[\Delta/T]}}, \quad (12)$$

where phenomenological parameters  $A$  and  $B$  are temperature independent but show a strong dependence on disorder. While the parameter  $A$  is the sheet conductance extrapolated to zero temperature, the disorder dependence of parameter  $B$  may be expressed in terms of the two microscopic parameters  $c$  and  $a$ :

$$\begin{aligned} B &\equiv \left(\frac{D}{D_0}\right)_0 \frac{2}{\pi} \left(\frac{12\pi}{5}\right)^{1/2} \left(\frac{4\Delta}{J}\right)^{3/2} t k_F \\ &= \frac{2c}{1 + \sqrt{1 + 4R_0^2/a^2}}, \end{aligned} \quad (13)$$

where

$$c \equiv \left(\frac{\Delta}{J}\right)^{3/2} \left(\frac{48t^2 k_F^2}{5\pi}\right)^{1/2}. \quad (14)$$

The data presented here is for a single film prepared with an initial  $R_0 \approx 6$  k $\Omega$ . Disorder was consequently increased in incremental stages up to 180 k $\Omega$  by annealing to temperatures above 150 K [14]. Additional samples were grown at intermediate disorder and measured to check reproducibility.

Figure 1 shows the conductivity data for the same sample at two different stages of anneal with corresponding disorder parameters  $R_0 = 17\,573$   $\Omega$  and 63 903  $\Omega$ . The solid lines are regression fits to the expression (12), where  $A$  and  $B$  are taken as fitting parameters and  $\Delta = 16$  K is the spin-wave gap. The fits are sensitive to the parameters  $A$  and  $B$  but relatively insensitive to  $\Delta$ . We find that  $\Delta = 16 \pm 4$  K provides good fittings in the whole range of disorder (from 6 to 180 k $\Omega$ ).

Figure 2 shows the dependence of the parameter  $B$  on the disorder strength  $R_0$  (open squares) and a theoretical fit (solid line) using Eq. (13), where  $c$  and  $a$  are fitting parameters. The

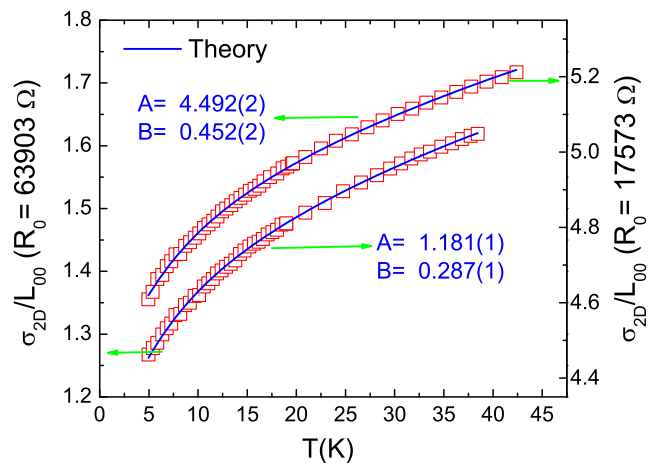


FIG. 1. (Color online) The temperature-dependent normalized conductivity (open squares) for two samples with the indicated disorder strengths of  $R_0 = 17\,573$   $\Omega$  and 63 903  $\Omega$  show good agreement with theory (solid lines). The fitting parameters  $A$  and  $B$  are indicated for each curve, with the error in the least significant digit indicated in parentheses.

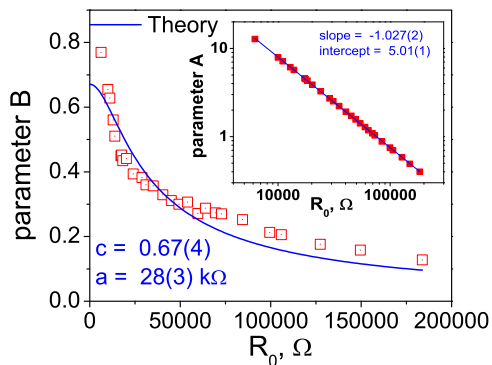


FIG. 2. (Color online) Dependence of the fitting parameters  $B$  and  $A$  (inset) on disorder  $R_0$  for  $\Delta = 16$  K. The fitting parameters are indicated for each curve, with the error in the least significant digit indicated in parentheses. Each of the 26 points in the main panel and in the inset represents a two-parameter fit ( $A$  and  $B$ ) to 26 temperature sweeps at different stages of disorder, two of which are shown in Fig. 1.

solid line for this two-parameter fit is drawn for the best-fit values  $c = 0.67 \pm 0.04$  and  $a = 28 \pm 3$  k $\Omega$ . We note that the fit is of reasonable quality over most of the disorder range, except for the film with the least disorder ( $R_0 = 6$  k $\Omega$ ) where  $B = 0.77$ , somewhat above the saturated value  $B = c = 0.67$  evaluated from Eq. (13) at  $R_0 = 0$ . Using higher values of  $c$  (e.g.,  $c = 0.8$ ) and lower values of  $a$  (e.g.,  $a = 22$  k $\Omega$ ) improves the fit at low disorder strengths but increases the discrepancy at higher disorder strengths.

Each of the 26 data points in Fig. 2 corresponds to a temperature sweep for 26 different stages of disorder as measured by  $R_0$ . Each sweep, two of which are shown in Fig. 1, gives two values for the  $A$  and  $B$  parameters. Accordingly, there are 26 values for  $A$  and  $B$  which are used in Fig. 2 to determine the two parameters  $a$  and  $c$ .

By substituting the Fermi energy for bulk Mn [24], a thickness  $t = 2$  nm, known to 20% accuracy, together with the best-fit value for  $c$  into Eq. (14), we calculate the value  $\bar{J} = 320 \pm 93$  K. Gao *et al.* [25] performed inelastic scanning tunneling spectroscopy (ISTS) on thin Mn films and reported  $\Delta$  in the range from 25 to 55 K (in satisfactory agreement with our value  $\Delta = 16 \pm 4$  K mentioned above) and spin-wave velocity  $A_s = 160 \pm 10$  meV  $\text{\AA}$ .

To make a closer connection between our transport measurements and the tunneling measurement results of Gao *et al.* [25], we note first that our  $\bar{J}$ , describing coupling of spin waves to particle-hole pairs written in terms of Pauli matrices, is smaller by a factor of 4 than the exchange interaction  $J$  in a Heisenberg Hamiltonian  $H = -(1/2) \sum_{i,j} J S_i S_j$ . In addition, a more accurate dispersion relation that takes into account the lattice structure relates the spin-wave velocity  $A_s$  to the Heisenberg  $J$  by  $A_s/a_0 = c_s J$ , where  $a_0$  is the

lattice spacing ( $a_0 \approx 1.8$   $\text{\AA}$  for our samples) and  $c_s$  is a dimensionless lattice parameter of order unity [26]. Thus the experimentally measured  $A_s$  of Gao *et al.* [25] corresponds to  $J = 1031 \pm 70$  K, while the theoretical fit corresponds to  $J = 4\bar{J} = 1280 \pm 360$  K. The results of the two experiments for the spin-wave gap and the exchange constant thus agree quite well.

Since the temperature-dependent correction  $B/\sqrt{\sinh(\Delta/T)}$  of Eq. (12) is small compared to the parameter  $A$ , we can write  $\sigma_{\square} \approx 1/R_0$  so that Eq. (12) reduces to the expression  $A \approx 1/L_{00} R_0$ . The logarithmic plot derived by taking the logarithm of both sides of this approximation is shown in the inset of Fig. 2. The slope of  $-1$  confirms the linear dependence of  $A$  on  $1/R_0$ , and the intercept of 5.01 ( $10^{5.01} \approx 102$  k $\Omega$ ) is within 20% of the expected theoretical value  $L_{00} = e^2/\pi h = 81$  k $\Omega$  for the normalization constant. Accordingly, the conductivity corrections in Eq. (12) are small compared to the zero temperature conductivity, and the normalization constant  $L_{00}$  for the conductivity is close to the expected theoretical value.

Using Eq. (11) and the obtained value for  $a \approx 28$  k $\Omega$  we can compare the dephasing length ( $L_{\phi}$ ) with the thickness ( $t \approx 2$  nm) at 16 K. For the sample with  $R_0 = 63903$   $\Omega$  the ratio  $L_{\phi}/t \approx 0.5$ , and for the sample with  $R_0 = 17573$   $\Omega$ ,  $L_{\phi}/t \approx 2$ . The latter estimate assumes no spin polarization, while a full polarization would imply  $L_{\phi}/t \approx 1$ . Thus  $L_{\phi}$  is smaller than or close to the thickness of the film, which keeps the film in the three-dimensional regime for almost all temperatures and disorder strengths considered.

## V. CONCLUSION

We have performed *in situ* transport measurements on ultrathin Mn films, systematically varying the disorder [ $R_0 = R_{xx}(T = 5$  K)]. The obtained data are analyzed within a weak-localization theory in 3D generalized to strong disorder. In the temperature range considered, inelastic scattering off antiferromagnetic spin waves is found to be strong, giving rise to a dephasing length shorter than the film thickness, which places these systems into the 3D regime. The obtained values for the spin-wave parameters in our transport measurements are close to the ones measured by Gao *et al.* [25] using ISTS. This good agreement further corroborates our finding of a 3D weak-localization correction at strong disorder that has not been previously identified.

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