Theory of Kondo suppression of spin polarization in nonlocal spin valves

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We theoretically analyze contributions from the Kondo effect to the spin polarization and spin diffusion length in all-metal nonlocal spin valves. Interdiffusion of ferromagnetic atoms into the normal metal layer creates a region in which Kondo physics plays a significant role, giving discrepancies between experiment and existing theory. We start from a simple model and construct a modified spin drift-diffusion equation which clearly demonstrates how the Kondo physics not only suppresses the electrical conductivity but even more strongly reduces the spin diffusion length. We also present an explicit expression for the suppression of spin polarization due to Kondo physics in an illustrative regime. We compare this theory to previous experimental data to extract an estimate of the Elliot-Yafet probability for Kondo spin flip scattering of 0.7 ± 0.4, in good agreement with the value of 2/3 derived in the original theory of Kondo.

I. INTRODUCTION

Pure spin currents, devoid of charge current flow, are now routinely generated in metals-based systems [1] via a number of techniques, including the use of thermal gradients [2], the spin Hall effect [3], spin pumping [4], and nonlocal spin injection [5], each method providing a unique insight into spin relaxation. In particular, the ability to separate charge and spin currents using the nonlocal spin valve [5, 6], thereby circumventing difficulties interpreting ‘local’ spin valve measurements, makes it one of the most unambiguous techniques for probing spin transport. This geometry is especially useful at the nanoscale, where isolating the factors affecting spin accumulation, diffusion, and relaxation, both within the bulk and across interfaces, represents a pressing problem [7–15]. Indeed, examining the role of specific defects in relaxing spins in metals at this length scale – including interfaces, grain boundaries, and magnetic and highly spin-orbit coupled impurities – will be critical for realizing future low resistance-area-product spintronic devices, e.g. current perpendicular-to-plane giant magnetoresistance sensors [16].

A nonlocal spin valve consisting of a normal metal channel connected by two ferromagnetic contacts is illustrated in Fig. 1(a): the injected current $I_{21}$ generates a spin accumulation at the interface between the nonmagnet and ferromagnet (Lead 2). This accumulation diffuses in both directions down the channel causing a pure spin current to flow towards Lead 1, which decays on a characteristic spin diffusion length, $l_{sN}$. The remaining spin population reaching Lead 3 generates a nonlocal voltage difference $V_{34}$ between the ferromagnetic contact and channel and therefore a nonlocal resistance, $R_{NL} = \Delta V_{34}/I_{23}$. The sign of this resistance depends on the relative orientation of the two ferromagnets, and so by applying a magnetic field to alternate the ferromagnetic contact magnetization from parallel to antiparallel, a nonlocal spin signal, $\Delta R_{NL}$, is measured, directly related to the magnitude of the spin accumulation under the contact.

In relatively simple all-metal nonlocal spin valves (e.g., Ni$_{80}$Fe$_{20}$/Cu) that are fabricated from nominally high-purity materials, the standard theory of spin drift-diffusion developed by Valet and Fert [17], combined with the Elliott-Yafet spin relaxation mechanism [18–20], which predominates in light metals, dictates that $l_{sN}^2$, the spin accumulation, and therefore $\Delta R_{NL}$ should monotonically increase as temperature $T$ decreases. Surprisingly, however, $\Delta R_{NL}$ is widely found to anomalously decrease at low $T$ in Ni$_{80}$Fe$_{20}$/Cu, Fe/Cu and Co/Cu nonlocal spin valves [21–30], even when the resistivity of the normal metal and the ferromagnet, $\rho_N(T)$ and $\rho_F(T)$, are found to continuously decrease on cooling.

Consensus is emerging that this unexpected reduction of $\Delta R_{NL}$ at low $T$ is due to spin relaxation at dilute magnetic impurities [26–28], with recent results demonstrating that a manifestation of the Kondo effect is at the heart of the suppression [29, 30]. The Kondo effect [31] arises in metals with dilute magnetic impurities, as a result of $s$-$d$ exchange between the conduction electrons and virtual bound impurity states. This exchange results in an additional higher order contribution to the scattering cross section, proportional to log $T$, which can dominate in otherwise highly pure metals at low $T$. In charge transport, the classic signature of the Kondo effect is an increase in the conduction electron scattering rate at low $T$, resulting in a minimum in resistance (maximum in conductance) and a logarithmic increase in $\rho(T)$ about a characteristic temperature $T_K$ [32]. Similarly, for spin transport the additional (spin-flip) scattering was recently found to efficiently relax the spin accumulation, suppressing $\Delta R_{NL}$ with...
what is also observed to be a log \( T \) dependence [29]. This occurs even for nonmagnetic channels that are largely impurity-free throughout the bulk, due to inevitable interdiffusion at the ferromagnet/nonmagnet interface. The situation is schematically depicted in Fig. 1(b), where interdiffusion creates a region with ‘high’ levels of ferromagnetic impurities (on average \( \approx 100’\)s \( \mu \text{mol/mol} \) [47]) which rapidly relax the injected spins at the interface, reducing the effective polarization of the bias current [29]. To this point, a direct quantitative link has already been established between the degree of interdiffusion and magnitude of Kondo suppression in nonlocal spin polarization near the interface due to the interdiffused ferromagnetic atoms over a characteristic length scale \( d \), which we term the Kondo region.

![Diagram](a) Geometry of a nonlocal spin valve, consisting of a normal metal (NM) channel and two ferromagnetic (FM) contacts. When an electrical current \( I_{21} \) is applied (blue line), a nonequilibrium spin accumulation develops, giving rise to a finite voltage difference \( \Delta V_{34} \) and nonlocal resistance \( R_{NL} = \Delta V_{34}/I_{21} \). (b) Schematic of the model used to determine the suppression of spin polarization at a ferromagnet/nonmagnet interface (\( z = 0 \)) due to interdiffusion ferromagnetic atoms over a characteristic length scale \( d \), which we term the Kondo region.

The paper is organized as follows. In Sec. II, we develop a theory describing suppression of the spin polarization at the interface. We first present the theory without derivation in Sec. II A and then we demonstrate that the spin polarization at the interface has a maximum at a finite temperature. In Sec. III, we compare the theory to our experimental data [30]. In Sec. IV, we present mathematical details which are referred to in Sec. II. Finally, in Sec. V we summarize the paper.

II. INTERFACIAL KONDO EFFECT

A. Modification of the electrical conductivity and the spin diffusion length due to the Kondo effect

Starting from antiferromagnetic exchange coupling between conduction electrons and dilute magnetic impurities, Kondo [31] showed that electrical conductivity in metals is suppressed at low temperature. This is equivalent to suppression of the momentum relaxation time:

\[
\frac{1}{\tau_N} = \frac{1}{\tau_N} + \frac{1}{\tau_K},
\]

where \( \tau_N \) is the momentum relaxation time without dilute magnetic impurities in the normal metal at the Fermi level and \( \tau_N \) is the modified momentum relaxation time in the presence of the Kondo effect. \( \tau_K^{df} \) is the effective Kondo relaxation time, for which the explicit expression is given below in Eq. (29). \( \tau_K^{df} \) has a logarithmic temperature dependence, and thus it can be comparable to or even dominate \( \tau_N \) at low temperature for very low impurity concentrations.

In addition to suppressing the scattering time (increasing the scattering rate), as in Eq. (1), the dilute magnetic impurities also suppress the spin relaxation time \( \tau_K^{sf} \):

\[
\frac{1}{\tau_{N}^{sf}} = \frac{1}{\tau_{N}^{sf}} + \frac{\eta}{\tau_K^{sf}},
\]

where \( \tau_{N}^{sf} \) is the modified spin relaxation time due to the Kondo effect. Here, \( \eta \) is the spin-flip probability during each Kondo scattering event. The proportionality between the change in the momentum relaxation rate (\( 1/\tau_K^{df} \)) and the spin relaxation rate (\( \eta/\tau_K^{sf} \)) is similar to that found for the Elliot-Yafet scattering mechanism. For Elliot-Yafet scattering, the contribution to the spin-flip scattering rate is given by \( 1/\tau \) where \( 1/\tau \) is the contribution to the momentum scattering rate and \( \beta \) is the Elliot-Yafet parameter. Thus, \( \eta \) is the inverse of the Elliot-Yafet parameter for spin relaxation from Kondo impurities. The value of \( \eta \) is determined by the geometry of the Fermi surface. For the spherical Fermi surfaces that we consider here, \( \eta = 2/3 \), as shown in Sec. IV. We note that in Ref. [28], the spin-flip probability is claimed to be around 0.3 based on a semiclassical argument [35]. Strictly, however, the...
semiclassical argument does not apply for the higher order interactions giving rise to the Kondo physics.

One remark on our notation is in order. Generally speaking \(\tau_N\) and \(\tau_N^{\text{sf}}\) are \(k\)-dependent functions. But here, we drop the \(k\) dependence and take the values only at the Fermi level for simplicity. In Sec. IV, we restore the \(k\) dependence for the derivation. In particular, \(\tau_N(k_F^N)\) and \(\tau_N^{\text{sf}}(k_F^N)\) in Sec. IV are respectively \(\tau_N\) and \(\tau_N^{\text{sf}}\) here, where \(k_F^N\) is the Fermi wavevector in the normal metal.

The changes in the relaxation times above imply changes in the electrical conductivity and the spin diffusion length. In Sec. IV, we show that the Valet-Fert theory for the spin drift-diffusion equation still holds even in the presence of dilute magnetic impurities, once we impose Eqs. (1) and (2). The Valet-Fert theory provides links between quantities in the Boltzmann equation (such as relaxation times) and quantities in the drift-diffusion equation (such as the electrical conductivity and spin diffusion length) as given below in Eqs. (19) and (20). Up to first order in the Kondo rate, the modified conductivity and spin diffusion length are

\[
\tilde{\sigma}_N = \sigma_N \left(1 - \frac{\tau_N}{\tau_N^{\text{eff}}} \right), \tag{3}
\]

\[
\tilde{l}_N = l_N^2 \left(1 - \frac{\tau_N + \eta \tau_N^{\text{eff}}}{\tau_E^{\text{eff}}} \right). \tag{4}
\]

Equation (3) corresponds to Kondo’s original work, i.e. the conductivity reduction due to the Kondo effect. Equation (4) is the spin counterpart of the original Kondo effect, a central aspect of this paper.

At this point it is worth noting that the Kondo effect can affect the spin diffusion length much more dramatically than it does the conductivity, since \((\tau_N^{\text{sf}})^{-1}\) is usually much smaller than \(\tau_N^{-1}\). For example, \(\tau_N^{\text{sf}}/\tau_N \approx 10^3\) in Ref. [1]. Thus it is possible that \(1 - \tilde{l}_N/l_N^2\) is noticeable even though \(1 - \tilde{\sigma}_N/\sigma_N\) is negligible.

### B. Suppression of the spin polarization at the interface

We now solve the spin drift-diffusion equation with the modified quantities in Eqs. (3) and (4) at the interface. Our model is illustrated in Fig. 1(b). We consider a ferromagnet \((z < 0)\)/nonmagnet \((z > 0)\) interface at \(z = 0\). Near the interface, interdiffused ferromagnetic atoms create a region in which dilute magnetic impurities are present. For illustration, we assume that the impurity concentration is constant over \(0 < z < d\) and suddenly drops to zero at \(z = d\). We term the region \(0 < z < d\) the Kondo region.

The spin drift-diffusion equation is given by the set of equations below [17].

\[
\frac{e}{\sigma(z)} \partial_z j_z(z) = \frac{\mu_z(z) - \mu_{-z}(z)}{l_z^2(z)}, \tag{5}
\]

\[
\partial_z \mu_z(z) = \frac{e}{\sigma_d(z)} j_z(z), \tag{6}
\]

where \(s = \pm\) denotes the spin majority and minority bands, \(e\) is the electron charge, \(j_z(z), \mu_z(z), \sigma(z), \) and \(l_z(z)\) are respectively the current expectation value, the chemical potential, the electrical conductivity, and the spin diffusion length of the spin \(s\) band at position \(z\). \(\sigma, \mu, \) and \(l_z\) are parameters treated as position independent in most cases. We explicitly retain the position dependence to emphasize the dependence of the parameters on the regions; \(z < 0, 0 < z < d,\) and \(z > d\). More explicitly, the parameters in each region are given by

\[
(\sigma(z), l_z(z)) = \begin{cases} 
(\sigma_{x,F}, l_{x,F}) & \text{for } z < 0, \\
(\sigma_N, l_N) & \text{for } 0 < z < d, \\
(\sigma_d, l_d) & \text{for } z > d.
\end{cases} \tag{7}
\]

Here the subscripts \(N\) and \(F\) refer to the normal metal and the ferromagnetic metal and the tildes refer to the Kondo region [48]. Since the physical parameters in the normal metal do not have spin dependence, we drop the subscript \(s\) for \(z > 0\).

The general solutions of Eqs. (5) and (6) are obtained in Ref. [17]. The spin accumulation \(\mu_z(z) - \mu_{-z}(z)\) decays exponentially over the effective spin diffusion length defined by \((l_z^{\text{eff}})^2 = l_z^2 + l_{x,F}^2\) for \(z < 0\), \((l_N^{\text{eff}})^2 = 2l_N^2\) for \(0 < z < d\), and \((l_d^{\text{eff}})^2 = 2l_d^2\) for \(z > d\). To match the experimental situation of Refs. [29] and [30], we assume transparent interfaces at \(z = 0\) and \(z = d\) where the spin chemical potential and the currents are continuous. The solutions of Eqs. (5) and (6) in the given situation are obtained in Appendix A. Here, we simply present spatial profiles of the spin accumulation \(\mu_z(z) - \mu_{-z}(z)\) and the spin current \(j_z(z) - j_{-z}(z)\) for a set of parameters in Fig. 2. For clear presentation, we scale the quantities. In this case, \(\mu_z(z) - \mu_{-z}(z)\) is normalized to its \(z = 0\) value, and \(j_z(z) - j_{-z}(z)\) to \(j_{\text{app}}/2.5\). Here \(j_{\text{app}} = j_z(z) + j_{-z}(z)\) is the applied charge current which is independent of position due to the conservation of electrical charge. The factor of 2.5 is simply to allow both quantities to be plotted on the same scale.

Figure 2 clearly shows that there is suppression of the spin polarization due to the Kondo region. Since there are no magnetic impurities for \(z > d\), relaxation rates for \(z > d\) are the same with and without the Kondo region. Thus, the spin accumulation calculated at \(z = d\) indicates the suppression of \(\Delta R_{NL}\) due to the Kondo effect. To quantify this suppression, we analytically evaluate the ratio of the accumulation at \(d\) in the presence and absence of a finite impurity concentration in the Kondo region (i.e. in the limit \(\tau_N^{\text{eff}} \to \infty\)). Here we compute the following expression up to \(O((\tau_N^{\text{eff}})^{-1})\):

\[
\kappa = \frac{\lim_{\tau_N^{\text{eff}} \to \infty} [\mu_z(d) - \mu_{-z}(d)]}{\lim_{\tau_N^{\text{eff}} \to \infty} [\alpha(d)]} = \frac{\alpha(d)}{\lim_{\tau_N^{\text{eff}} \to \infty} [\alpha(d)]}, \tag{8}
\]

where \(\alpha(z)\) is defined as the current polarization, \(\alpha(z) = [j_z(z) - j_{-z}(z)]/j_{\text{app}}\) [49]. At this point it is worth emphasising that, since it is a quantitative indication of the strength of Kondo suppression, the suppression ratio \(\kappa\) represents a key parameter of this work. Furthermore, as \(\alpha\) is directly measurable, \(\kappa\) uniquely represents an experimentally accessible spin transport parameter with which to compare theory and measurement. In evaluating Eq. (8) we retain only terms up to \(O(d)\), assuming that \(d\) is much shorter than the effective spin


III. DETAILED COMPARISON WITH EXPERIMENTAL DATA

The suppression of the spin polarization $\alpha$ [quantified by $1 - \kappa$ in Eq. (9)] has been experimentally observed in nonlocal spin valves fabricated from a variety of miscible, moment-forming ferromagnet/nonmagnet pairings, e.g., Fe/Cu and Ni$_{80}$Fe$_{20}$/Cu [29]. Recently, through the use of thermal annealing to promote interdiffusion, we have also shown fine control over $\kappa$ in Fe/Cu nonlocal spin valves, and directly correlated its magnitude to the Fe/Cu interdiffusion length, $\lambda_{\text{Fe}}$ [30]. Equation (9) quantitatively connects $\kappa(T)$ to measurable quantities for the first time, and so provides an ideal expression with which to compare to these experiments. In the current section, we examine the experimental magnitude and $T$ dependence of $\kappa$, while varying the annealing temperature, $T_A$, in order to tune the extent of the interfacial Kondo region. Through this analysis, and the use of Eq. (9), we extract an experimental value for the Elliot-Yafet probability for Kondo spin-flip scattering $\eta$, demonstrating good agreement between the presented theory and experimental results.

In nonlocal spin valve measurements, where ferromagnetic contacts are separated by a distance $L$, $\alpha(T)$ is typically extracted by fitting $\Delta R_{\text{NL}}(L, T)$ to a one-dimensional model of nonlocal spin transport [39]. In this model $\alpha_{\text{eff}}$ enters as a boundary condition which principally determines the magnitude of $\Delta R_{\text{NL}}$ at fixed $L$. Provided that the Kondo region is small compared with the mesoscopic device length ($d < L$), Kondo depolarization then appears as an interfacial effect, and is manifest as a suppression of the measured $\alpha$ at low $T$, as quantified by $\kappa$ [Eq. (9)]. To account for this, we define an effective polarization, $\alpha_{\text{eff}} = \alpha(d)$, i.e., the observed current polarization in nonlocal spin valves with Kondo suppression present. This can be contrasted with the intrinsic polarization of the ferromagnet, $\alpha_{\text{FM}}$. In this context, $\kappa \approx \alpha_{\text{eff}}/\alpha_{\text{FM}}$, and so determining $\alpha_{\text{eff}}(T)$, through $\Delta R_{\text{NL}}(L)$, and $\alpha_{\text{FM}}(T)$ yields an experimental measure of $\kappa(T)$. In the following, we examine $\alpha_{\text{eff}}(T)$ obtained from annealed Fe/Cu nonlocal spin valves. (Details of experimental fabrication and measurement of these all-metallic nonlocal spin valves can be found in the original reports). We note that Fe/Cu represents an ideal choice of materials as Fe is miscible and moment forming in Cu [40], with a readily accessible $T_K = 30$ K [41].

To determine $\alpha_{\text{FM}}(T)$ we also measured $\Delta R_{\text{NL}}(L, T)$ in devices devoid of dilute impurity moments, and thus the Kondo effect. Two types of devices were tested along these lines: nonlocal spin valves fabricated from nonmagnets that do not support local moments, e.g., Al; and nonlocal spin valves that incorporate a thin interlayer (e.g., Al) between the ferromagnet and nonmagnet that suppresses interdiffusion and moment formation. In both types of device the normalized $\alpha_{\text{FM}}(T)$ is found to be monotonic and quantitatively similar, as shown in black squares in Fig. 3(a).

To avoid complications from potential interface resistance changes during annealing, as well as other inherent systematic errors between devices, we scale $\alpha_{\text{eff}}$ to $\alpha_{\text{FM}}(T)$ using the method discussed in Appendix B. The resulting normalized $\alpha_{\text{eff}}$ for various $T_A$ are shown in Fig. 3(a), with the corre-
In this equation, $\tau$ decreases with increasing measurement temperature $T$. For devices that do not support local moments $A$, the effect of interdiffusion, however, a suppression of $\alpha$ is observed at low $T$, the magnitude of which broadly increases with increasing $T_{\lambda}$. Consequently, $1 - \kappa(T)$ is largest at high $T_{\lambda}$ and decreases with increasing measurement $T$, as would be anticipated. Figure 4(a) shows $1 - \kappa(T)$ at $T = 5$ K for all samples, demonstrating this increase in magnitude with $T_{\lambda}$. Note Fig. 4 displays data for all eight measured device batches, including multiple sets at $T_{\lambda} = 200$ °C and 300 °C.

We now consider fitting the data of Fig. 3(b) using Eq. (9). In this expression $\rho_{\text{in}}(T)$, $\rho_{\text{in}}(T)$, and $\rho_{\text{in}}(T)$ are measured experimentally. $\tau_{\text{in}}^N$ is constrained to the literature value of 950 [42] — a value we have explicitly verified in high-conductivity nonlocal spin valve channels, where Kondo effects are negligible and phonon scattering dominates spin relaxation. We consider two expressions for $\rho_{\text{in}}(T)$, the Kondo model [31] [Eq. (10)] and the phenomenological formalism of Goldhaber-Gordon (G-G) [36] [Eq. (11)]:

$$\rho_{\text{in}}^K = \rho_{\text{in}}^K \left( 1 + 2N_0 J (T_{\text{m}}^2 + T_{\text{m}}^2)^{1/2} \ln \left( \frac{U}{k_B T_{\text{m}}} \right) \right)$$

$$\rho_{\text{in}}^{G-G} = \rho_{\text{in}}^{G-G} \left[ 1 + 2N_0 J (T_{\text{m}}^2 + T_{\text{m}}^2)^{1/2} \ln \left( \frac{U}{k_B T_{\text{m}}} \right) \right]$$

where $T_{\text{m}} = T_{\text{K}} / \sqrt{2^{1/2} - 1}$, $\rho_{\text{in}}^K = \tau_N^2 F_{\text{Fe}}^2 S(S + 1)/\tau_N^2 J^2 m/3\hbar n_{\text{Fe}}^0 e^2$ is the classical resistivity of dilute magnetic impurities, $U$ is the on-site Coulomb energy, $N_0$ is the density of states of each spin band, $J$ is the (negative) exchange parameter between the conduction electron and the magnetic impurities, and $s$ is the so-called G-G exponent which is typically taken to be 0.22 [36]. For the expression for $\rho_{\text{in}}^{G-G}$, $\mu_{\text{Fe}}^0$ is the Fermi level of the normal metal, $\tau_N^2$ is the average impurity concentration in Fe, $S$ is the spin angular momentum of the magnetic impurities, $n_{\text{Fe}}^0$ is the density of electrons, $m$ is the effective electron mass. Here we have adapted the generalized G-G model to give agreement with Kondo’s original theory. $T_{\text{m}}$ depends on the limits of the energy integration of Eqs. (24) and is typically taken to be either $T_{\text{K}}$, $U$ or $J$, depending on the theoretical treatment. In our case, the requirement that both models be equivalent at $T = T_{\text{K}}$ gives $T_{\text{K}} = k_B T_{\text{K}}^2 / U$. The Fermi energy of Cu, $\mu_{\text{Cu}}^0 = 7$ eV [43], is well known, as is $T_{\text{K}} = 30$ K from $\rho_{\text{in}}(T)$ and susceptibility measurements [41]. Furthermore, $J = 0.91$ eV for Fe/Cu has been experimentally measured via field-dependent magneto-resistance and magnetometry measurements [44]. From our own measurements of $\rho_{\text{in}}(T)$ in heavily doped Cu/Cu nanowires, we can establish $U \approx 0.86$ meV (10 K). Noting $\tau_N^2 / \tau_N \gg 1$, this leaves only the product $n_{\text{Fe}}^2 d$, i.e., the weighted total number of impurities in the Kondo region (per cross sectional area), as an unknown. Equation (2) is known to be valid only over a limited $T$ range about $T_{\text{K}}$, evolving to the value dictated by the classical scattering rate at $T \gg T_{\text{K}}$ and the unitary limit at $T \ll T_{\text{K}}$. Consequently, when using this model we restrict the $T$ range to only consider the transition region within the data, between approximately 10 K and 100 K. In addition to considering both of these models, we also compare to the cases where $T_{\text{K}}$ is allowed to be an unconstrained fitting parameter. The extracted $n_{\text{Fe}}^2 / \tau_N$ from each model (through a least mean square minimization of residuals) is shown in Fig. 4(b). We note that the individual parameters $\eta$, $C_{\text{Fe}}$, and $d$ remain otherwise inseparable. The solid lines in Figure 3(b) show fits of $1 - \kappa(T)$ using the G-G model with unconstrained $T_{\text{K}}$. In general, the overall magnitude and $T$ dependence is well captured for all $T_{\lambda}$. When $T_{\text{K}}$ is unconstrained we find $T_{\text{K}} = (44 \pm 36)$ K for the G-G model and $T_{\text{K}} = (22 \pm 9)$ K for the Kondo model, in good agreement with the literature value of $T_{\text{K}} = 30$ K. (All uncertainties in this paper are single standard deviations, the determination of which is discussed in Appendix D.) The behavior of these values reflects the logarithmic dependence
of $\rho_K$ on $T$, and so the difficulty in determining $T_K$, which is often a challenge in dilute-moment metallic systems. Despite the limited range of applicability, both expressions (i.e., for $\rho_K^{fK}$ and $\rho_K^{fG}$) give consistent results for $\eta\hat{C}_{Fe,d}$, reflecting the fact that these parameters only act to influence the magnitude of $\kappa$, and are thus relatively insensitive to the precise $T$ dependence of the data. Examining Fig. 4(b) we see that the total number of impurities increases on annealing, with dramatic changes occurring above $T_A \approx 300 \, ^\circ C$, in good agreement with our previous observations of the $T_A$ dependence of $\lambda_{Fe}$ in this system [30].

To place these values of $\eta\hat{C}_{Fe,d}$ in context, and to extract a value of $\eta$, we consider our previous work on interdiffusion in Fe/Cu nonlocal spin valves. Fe has finite solubility in Cu, with a limit of 2600 $\mu$mol/mol at room temperature [40] (based on the bulk equilibrium phase diagram), beyond which precipitation occurs, leading to phase-segregated clusters. In the following analysis we therefore assume that regions with $C_{Fe} > 2600 \, \mu$mol/mol do not contain isolated dilute moments, and so do not contribute to the Kondo effect. Through energy-dispersive x-ray spectroscopy (EDX) measurements in cross-sectional scanning transmission electron microscopy (STEM) we have previously shown that the interdiffusion profile in annealed Fe/Cu nonlocal spin valves follows $C_{Fe}(x) \approx [1 - \text{erf}(x/\lambda_{Fe})]/2$, and have quantitatively determined $\lambda_{Fe}(T_A)$ for our devices [30]. Using this expression for $C_{Fe}$, considering only the dilute Kondo region below $C_{Fe} < 2600 \, \mu$mol/mol in nonlocal spin valves, and assuming $L \gg \lambda_{Fe}$, yields a total number of impurity atoms [50] (per unit cross sectional area) in the Kondo region of $C_{Fe,d} = 560 \, \mu$mol/mol $\cdot \lambda_{Fe}$. Using this result and our previous measurements of $\lambda_{Fe}$, we are therefore able to estimate $\eta$. Duly extracted values of $\eta$ are shown in Fig. 4(c) as a function of $T_A$, with the average $\bar{\eta} = 0.7 \pm 0.4$ indicated by the solid horizontal line. We anticipate that the simplicity of the model in accounting for the precise dispersion of Fe at the Fe/Cu interface, in addition to variability in the precise degree of interdiffusion at intermediate $T_A$, likely accounts for the dispersion in the extracted values of $\eta$, particularly at $T_A = 200 \, ^\circ C$. This is also likely to be the cause of the unphysical value of $\eta > 1$ found at $T_A = 450 \, ^\circ C$ (note also the large random error). Despite the variation, we find good overall consistency between the experimentally determined values and $\eta = 2/3$ (dashed horizontal line), as calculated in the original work of Kondo [31]. Given the rather simplistic assumptions made, as well as the use of four independent experimental measurements in order to determine $\eta$, this result indicates good consistency between the model and experiment.

As a final point of discussion, we note that one of the most striking observations of Ref. [30] was the failure of the standard one-dimensional Valet-Fert model to describe the temperature and separation dependence of $\Delta R_{NL}$ at high $C_{Fe}$. It is important to highlight that, due to heavy interdiffusion, this situation involves not only a large $C_{Fe}$, but also a large gradient over relatively long distances. The conclusion from this should not be that the Valet-Fert theory itself is fundamentally incapable of describing the impact of Kondo relaxation on spin transport. Rather, it is the application of models, based on Valet-Fert theory but which, crucially, ignore Kondo physics, which fails to describe even qualitative observations. The results in this work suggest, in fact, that it is indeed possible for an (albeit modified) Valet-Fert approach to describe previous experimental results, when the precise distribution of local moments throughout the device is known.

IV. DERIVATIONS

In this section we present mathematical derivations of the core results in Sec. II A. First we start from the Boltzmann equation to derive the spin drift-diffusion equation at finite temperature. We take the approach developed by Valet and Fert [17]. Readers not familiar with details of the Valet-Fert theory may refer to Appendix C 1.

We start from the Boltzmann equation for a translation-invariant system in the two directions perpendicular to $z$. The distribution functions $f_{\pm}$ are a function of $z$ and $k$. At the
equilibrium, they are \[ f_i^0(z, k) = e^{-\beta \varepsilon_i(k) + \mu_0(z)} + 1 \] where \( \varepsilon_i(k) = \hbar^2 k^2 / 2m \) is the energy eigenvalue, \( k \) is the crystal momentum, \( k = |k| \), \( \mu_0(z) \) is the chemical potential at the equilibrium, \( m \) is the effective mass, \( \beta = 1 / k_B T \) is the inverse temperature, and \( k_B \) is the Boltzmann constant. When an electric field is applied along the \( \hat{z} \) direction, the distribution function has a small correction to its equilibrium function \( f_i(z, k) = f_i^0(z, k) + g_i(z, k) \) where \( g_i(z, k) \) is the small correction proportional to the electric field and \( s = \pm \). The linearized Boltzmann equation at steady state under the relaxation time approximation is

\[
\frac{\hbar k}{m} \partial_z g_s(z, k) - \frac{eE}{\hbar} \partial_z f_i^0(k) = -\frac{g_s(z, k) - g_s^0(z, k)}{\tau_s^c(z, k)} - \frac{g_s(z, k) - g_s^0(z, k)}{\tau_s^f(z, k)},
\]

(12)

where \( \tau_s^c \) and \( \tau_s^f \) are the relaxation times. \( \tau_s^c \) corresponds to spin-conserving processes \( s \to s \), and \( \tau_s^f \) corresponds to spin-flipping processes that gives the spin diffusion length. A two-channel model works well in materials with weak spin-orbit coupling, like Cu, where the different cartesian components of the spin are largely decoupled from one another. This allows us to neglect spin components perpendicular to the magnetization in the ferromagnet in our derivation. The sum of the spin-conserving and the spin-flipping rates gives the electrical conductivity described below. These relaxation times are assumed to be isotropic, that is, they only depend on the magnitude of \( k \). In general \( \tau_s^c \) is independent of \( s \) at the Fermi surface. However, since the Fermi wave vector depends on \( s \), \( \tau_s^c \) written as a function of \( k \) is \( s \) dependent. This is also true for the Kondo contribution [31]. The relaxation times \( \tau_N \) and \( \tau_N^0 \) that appear in Sec. II will be connected to these functions. \( g_s^0(z, k) \) is the angle-averaged \( g_s(z, k) \) over the Fermi surface with the constant magnitude \( |k| = k \), that is, \( g_s^0(z, k) = (1/4\pi) \int_{-\pi/k}^{\pi/k} d\Omega_k g_s(z, k) \) where \( \Omega_k \) is the solid angle of \( k \).

We make the approximation that the system is rotationally symmetric around the \( z \) axis. In this regime, \( g_s(z, k) \) can be expanded by the Legendre polynomials as \( g_s(z, k) = \sum_{n=0}^{\infty} g_s^{(n)}(z, k) P_n(\hat{k} \cdot \hat{z}), \) where \( P_n \) is the Legendre polynomial. Since the Legendre polynomials form an orthonormal set of polynomials, each coefficient satisfies the equation. Taking \( n = 0, 1 \) coefficients and neglecting the higher order contributions [17], Eq. (12) is equivalent to

\[
\frac{\hbar k}{3m} \partial_z g_{s}^{(1)}(z, k) = -\frac{g_{s}^{(0)}(z, k) - g_{s}^{(1)}(z, k)}{\tau_s^f(z, k)},
\]

(13)

\[
\frac{\hbar k}{m} \partial_z g_{s}^{(0)}(z, k) = -\frac{g_{s}^{(1)}(z, k)}{\tau_s^c(z, k)},
\]

(14)

where \( g_{s}^{(0)} = g_{s}^{0} - eE \partial_z f_i^0, 1 / \tau_s = 1 / \tau_s^c + 1 / \tau_s^f \) is the total scattering-out rate of a spin \( s \) state.

Equations (13) and (14) are the spin drift-diffusion equations that hold at \( T = 0 \). At \( T = 0 \), \( g_{s}^{(0)} \) and \( g_{s}^{(1)} \) evaluated at the Fermi surface are respectively assigned to the chemical potential and the current, with proper prefactors [17]. Therefore, Eqs. (13) and (14) provide closed solutions for these physical quantities. However, this association is not exact at finite temperature. For \( T > 0 \), a physical quantity is not given by a value at the Fermi surface, but is given after integrating over \( k \), considering \( T \) dependence of \( f_i^0(k) \).

Although the temperature dependence of \( f_i^0 \) is very complicated, the Sommerfeld expansion formula in Appendix C2 allows substantial simplification. The Sommerfeld expansion formula is an expression for integrals including the Fermi-Dirac distribution at low temperature. In the Kondo regime, \( k_B T \ll \mu_0 \) satisfies the criterion. Neglecting \( O(T^2) \), the spin density \( n_s \) for each \( s \) band (or the spin chemical potential with an additional factor as shown below) is given by

\[
n_s(z) = \frac{e}{V} \sum_k f_i(z, k) = \frac{e}{2\pi^2} \int dk k^2 \tilde{g}_s^{(0)}(z, k) + \frac{m\sqrt{2m_0(z)}\mu_0(z)e}{3\pi^2\hbar^3}.
\]

(15)

We use here the Sommerfeld expansion formula Eq. (C7) for low temperature \( k_B T \ll \mu_0 \). Similarly, the current density \( j_s(z) \) for \( s \) band is

\[
j_s(z) = -\frac{e}{V} \sum_k \frac{\hbar k}{m} f_i(z, k) = -\frac{eh}{6\pi^2 m} \int dk k^2 \tilde{g}_s^{(1)}(z, k).
\]

(16)

Assuming that \( \mu_0(z) \) is constant in space in each region (sudden changes at the boundaries can be taken into account by matching boundary conditions), integration of Eqs. (13) and (14) with the weighting factors \( k^2 \) and \( k \) gives respectively

\[
\partial_z \tilde{j}_s(z) = \frac{e}{2\pi^2} \int dk k^2 \tilde{g}_s^{(0)}(z, k) - \tilde{g}_s^{(1)}(z, k),
\]

(17)

\[
\partial_z \tilde{n}_s(z) = -\frac{em}{2\pi^2 \hbar} \int dk k^2 \tilde{g}_s^{(1)}(z, k).
\]

(18)

Equations (17) and (18) provide a generalized drift-diffusion equation at low temperature.

We first briefly show that Eqs. (17) and (18) become the conventional spin drift-diffusion equations without the Kondo effect. That is, Valet-Fert theory holds up to \( O(T) \), if there are no magnetic impurities. Without an electric field, \( \tilde{g}_s^{(0)} \) and \( \tilde{g}_s^{(1)} \) are zero. This observation implies that \( \tilde{g}_s^{(0)} \) and \( \tilde{g}_s^{(1)} \) are proportional to \( \partial_z f_i^0 \). By the Sommerfeld expansion formula, \( \partial_z f_i^0 \) can be replaced by \( -\partial_z \mu_0 - \varepsilon \), neglecting \( O(T^2) \). Thus, the integrations Eqs. (17) and (18) are nothing but evaluations at the Fermi surface. Therefore, Eqs. (17) and (18) are equivalent to Eqs. (13) and (14) up to \( O(T) \).

Now we connect the quantities that appear in Eqs. (13) and (14) to physical quantities. First we define the electrical conductivity \( \sigma_s(z) \) and the spin diffusion length \( l_s(z) \) for each spin band \( s \), which are respectively given by

\[
\sigma_s(z) = \frac{e^2(k_f^s)^3}{6\pi^2 m} \tau_s(z, k_f^s),
\]

(19)

\[
l_s^2(z) = \frac{\hbar^2(k_f^s)^2}{3m^2} \tau_s(z, k_f^s) \tau_s^f(z, k_f^s).
\]

(20)
where \( k_F^e = \sqrt{2m_F m/\hbar^2} \) is the Fermi wave vector. Here the electrical conductivity is equivalent to the Drude conductivity \( \sigma = n_e e^2 (\tau_e(k_F^e)^2)/m \), where \( n_e = (k_F^e)^2/2\pi^2 \) is \( n_e \) without an electric field. Similarly, the spin diffusion length is related to the diffusion constant by \( L_s^2 = D_s \tau_s^e(k_F^e) \) where \( D_s = h^2(k_F^e)^2 \tau_s(z, k_F^e)/3m^2 \). Next, we define the electrochemical potential \( \mu_e = (2\pi^2 k^2 e^2 m)^{1/2} \). The factor arises from the ratio between the charge and \( (e/V)^N \) in Eq. (15). With these definitions, Eqs. (13) and (14) become equivalent to Eqs. (5) and (6).

The situation changes in the presence of dilute magnetic impurities. We show that Eqs. (5) and (6) are still valid after replacement of Eqs. (3) and (4) [or equivalently Eqs. (1) and (2)] and give explicit expressions for \( \tau_s^\sigma \) in particular regimes. Since the Kondo effect occurs in the normal metal, we use the subscript \( N \) but drop the spin-dependent subscript \( s \). That is, \( \tau_N(z, k) \) and \( \tau_s^N(z, k) \) are the relaxation times in the normal metal, which are spin independent. \( \tau_N \) and \( \tau_s^N \) that appear in Sec. II are those evaluated at the Fermi level \( k = k_F^e \). In the presence of dilute magnetic impurities, additional relaxation rates arise due to the impurities. We denote these by the subscript \( K \). In the Boltzmann equation Eq. (12), the relaxation times change by

\[
\frac{1}{\tau_N(z, k)} = \frac{1}{\tau_s^N(z, k)} + \frac{1}{\tau_k^N(z, k)} \quad \text{(21)}
\]

\[
\frac{1}{\tau_K(z, k)} = \frac{1}{\tau_s^N(z, k)} + \frac{1}{\tau_k^N(z, k)} \quad \text{(22)}
\]

where \( \tau_s^N \) and \( \tau_k^N \) are the modified relaxation times due to the Kondo effect. Kondo [31] computed explicitly the total scattering rate change \( \tau_k^N = (\tau_s^N)^{-1} + (\tau_k^N)^{-1} \) given by

\[
\frac{1}{\tau_k(z, k)} = \frac{2n_F^0 (S + 1) C N_0^2 J^2}{3\hbar} [1 + 2J\gamma(e)], \quad \text{(23)}
\]

\[
\gamma(e) = \frac{1}{V} \sum_k \frac{n_F^0(k)}{\varepsilon(k) - e} \quad \text{(24)}
\]

where \( S \) is the spin angular momentum of the magnetic impurities, \( N_0 = mk_F^e \/2\pi^2 \hbar^2 \) is the density of states of each spin band, \( J \) is the (negative) exchange parameter between the conduction electron and the magnetic impurities, and \( \hat{C} \) is the average impurity concentration, which is the density of impurities divided by the density of electrons \( 2n_F^0 = (k_F^e)^2/2\pi^2 \). The units of \( J \) are \( J \cdot \text{m}^{-3} \) to make \( N_0 J \) dimensionless. To be explicit, we set the Kondo Hamiltonian to be \( H_K = -(4J/V) \sum_k \varepsilon(k) \hbar \{ \Psi_k^d \sigma \Psi_{k'}^a \} \Psi_{k'}^d \hbar \{ \Psi_{k'}^a \sigma \Psi_{k}^d \} \), where \( \Psi_k^d \) and \( \Psi_{k'}^d \) are respectively the electron creation operator of conduction electrons with momentum \( k \) and electrons in the impurity state \( d \) and \( \sigma \) is the Pauli matrix.

Since the Kondo theory is a perturbation theory, its contribution to the spin-flip rate \( (\tau_s^N)^{-1} \) is likely to be proportional to Eq. (23) as Kondo showed [31]. We introduce \( \eta \), the spin-flip probability during each Kondo scattering event by

\[
\frac{1}{\tau_k^N(z, k)} = \eta \frac{1}{\tau_k(z, k)}. \quad \text{(25)}
\]

The value of \( \eta \) is determined by the Fermi surface geometry. For a spherical Fermi surface that we use here, Kondo [31] showed that \( (\tau_k^N)^{-1} \) is twice \( (\tau_k^N)^{-1} \) [51]. This indicates that \( \eta = 2/3 \) for this case.

The low temperature behavior of \( \tau_k(z, k) \) requires careful treatment. Since \( \gamma(e) \) diverges at the Fermi level, naive application of the Sommerfeld expansion gives divergences. Although the integrals in Eqs. (17) and (18) seem surprisingly difficult to perform without the Sommerfeld expansion, a low temperature approximation allows it. In Appendix C, we slightly generalize Kondo’s approach to extract the logarithmic dependence of the Kondo resistivity to show that the following replacement is valid under the integration over the energy.

\[
\gamma(e) \partial e \phi^0 \rightarrow N_0 \ln \frac{k_F^e T}{\mu_0^N} [\delta(e - \mu_0^N)], \quad \text{(26)}
\]

giving rise to a \( \ln T \) contribution. With this rule, Eqs. (5) and (6) still hold under the following replacement.

\[
\frac{1}{\tau_N(z, k)} \rightarrow \frac{1}{\tau_s^N(z, k)} \equiv \frac{1}{\tau_k^N(z, k)} + \frac{1}{\tau_k^N(z, k)} \quad \text{(27)}
\]

\[
\frac{1}{\tau_k(z, k)} \rightarrow \frac{1}{\tau_k(z, k)} \equiv \frac{1}{\tau_k(z, k)} + \frac{\eta}{\tau_k(z, k)} \quad \text{(28)}
\]

which are nothing but Eqs. (1) and (2). Here

\[
\frac{1}{\tau_k^N} = \frac{2n_F^0 S (S + 1) C N_0^2 J^2}{3\hbar} \left( 1 + 2N_0 J \ln \frac{k_F^e T}{\mu_0^N} \right). \quad \text{(29)}
\]

V. SUMMARY

In order to take account for the Kondo effect in spin transport, we derive a modified drift-diffusion equation from the Boltzmann equation explicitly allowing for finite temperature. The complicated finite temperature theory is projected to a low temperature regime (compared to the Fermi temperature). We show that the Valet-Fert drift-diffusion equation holds both at finite \( T \) and in the presence of spin scattering from dilute magnetic impurities, once the electrical conductivity and spin diffusion length are renormalized as functions of temperature. This represents a useful result; as a consequence, dilute magnetic impurity scattering beyond the semiclassical limit can indeed be described in a simple Elliot-Yafet-like form with a direct proportionality between \( \tau_k^N \) and \( \tau_k \), as originally indicated by Kondo. The modified drift-diffusion equation has a remarkably compact form given the complexity of the higher order many-body interactions involved.

By solving the drift-diffusion equation for an illustrative regime, we show additional spin relaxation in the presence of the Kondo effect at a ferromagnet/nonmagnet interface. Kondo scattering is found to be highly efficient at spin relaxation, due to the high probability of spin flip (\( \eta = 2/3 \)) compared with other scattering mechanisms (c.f. \( \eta_{\text{phonon}} \approx 1/1000 \)). Since the spin-flip rate is much lower than the moment scattering rate in the absence of the Kondo effect,
such a high probability caused by the Kondo effect can significantly reduce the spin diffusion length, even when there is negligible change to the conductivity. This is confirmed experimentally by the large value of $\eta \approx 0.7$ observed, in good agreement with Kondo’s original work. We hope this, in addition to the explicit derivation of Eq. (4), further validates the semiclassical model of Ref. [28] in also determining the experimentally by the large value of $\eta$. This is confirmed experimentally extracted, as well as approximations regarding the precise distribution of magnetic moments within the Kondo region. Examining Fig. 4, one can see a weak dependence of $\eta$ on $T_A$. This is very likely due to such simplifications. Indeed, the possibilities of Fe segregation and cluster formation on annealing, dilute impurity migration to grain boundaries, and inter-moment correlations at high concentrations, as well as examining the precise phase equilibrium beyond the thermodynamic limit are entirely overlooked, and could greatly complicate the situation. Nevertheless, agreement between the simple model and experiment is highly satisfactory.

One observation worth mentioning is the form of Eq. (9), particularly the fact the signal suppression is linear in both $C_{Fe}$ and $d$. This clarifies one of the fundamental difficulties previously experienced within the field. That is, determining the precise location of the anomalous relaxation mechanism. Previous reports have stated relaxation occurring at the ferromagnet/nonmagnet interface (as we discuss here), throughout the channel [27, 28], or at surfaces [21, 26, 45], with similar magnitudes of Kondo suppression in each case. To first order it is the product $C_{Fe}d$ (total number of impurities per cross-sectional area) that determines suppression, and so similar magnitudes may be observed either due to a high-impurity-concentration narrow region (e.g., an interfacial effect), or an extended low concentration region (i.e., low doping levels throughout the channel itself). For the case where magnetic impurities extend throughout the channel, i.e., in the limit where $d \geq l_N$, the approximations made in obtaining Eq. (9) will no longer be appropriate. Instead separation-dependent measurements of $\Delta R_{NL}$ on mesoscopic lengthscales (i.e. comparable to $l_N$) will follow the standard non-local spin transport equations, now with the modified value of $l_N$ given by Eq. 4. For low impurity levels this results in comparable magnitudes of suppression to those seen here. Rather than serendipitous, it is entirely expected that both the interfacial effects discussed here and ‘contaminated’ channel devices observe similar signal contributions from Kondo effects.

This highlights the care that must be taken when fitting $\Delta R_{NL}(L, T)$ to resolve the contributions from interfacial (manifest in the extracted $a$) and bulk (manifest in $l_N$) Kondo relaxation, in the likely scenario where both cause $\Delta R_{NL}$ to be suppressed by comparable amounts.

Having now determined the theoretical and experimental $T$-dependence of Kondo spin scattering in nonlocal spin valves, this opens the path to using the Kondo effect to better understand magnetic and nonmagnetic impurity spin relaxation. In particular, the clearly identifiable $T$-dependence may now be used as a signature to quantitatively determine the contribution of dilute moments to relaxation in all-metal systems.

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Appendix A: Solution of the spin drift-diffusion equation

In Ref. [17], the general solutions for Eqs. (5) and (6) are given by

$$\begin{align*}
\mu_+(z) - \mu_-(z) &= \begin{cases}
A_F e^{i\eta z} & \text{for } z < 0, \\
\tilde{A}_F e^{i\eta z} + B_N e^{-i\eta z} & \text{for } 0 < z < d, \\
B_N e^{-i\eta z} & \text{for } z > d,
\end{cases} \\
\sigma_+(z)\mu_+(z) + \sigma_-(z)\mu_-(z) &= \begin{cases}
C_F + D_F & \text{for } z < 0, \\
\tilde{C}_F + D_N & \text{for } 0 < z < d, \\
C_N + D_N & \text{for } z > d.
\end{cases}
\end{align*}$$

(A1) (A2)

In this section we determine the coefficients satisfying the transparent boundary conditions

$$\begin{align*}
\mu_+(z = -0) = \mu_+(z = +0), \quad \mu_+(z = d - 0) = \mu_+(z = d + 0), \quad j_s(z = -0) = j_s(z = +0), \quad j_s(z = d - 0) = j_s(z = d + 0).
\end{align*}$$

(A3)
There are eight boundary conditions (note that \( s = \pm \)) although there are ten unknown coefficients. Therefore, two more conditions are required. The first one originates from a constant shift of the chemical potential. Since the drift-diffusion equation is invariant under a constant shift of the chemical potential, we can put \( D_N = 0 \) without any loss of generality. The second one originates from the homogeneity of the drift-diffusion equation. The drift-diffusion equation is invariant under multiplication by a constant factor to \( \mu_s \). The applied electrical current defined by

\[
e_{j_{\text{app}}} = e_j(z = -\infty) + e_j(z = -\infty) = C_F,
\]

is an experimentally controllable quantity that fixes the multiplication factor.

Now we apply the boundary conditions. Instead of applying the continuity of each functions, we can apply it with their independent linear combinations. Note that \( \mu_+(z) - \mu_-(z) \) is already given above and \( j_+(z) - j_-(z) \) is nothing but the derivative of \( \sigma_+(z)\mu_+(z) + \sigma_-(z)\mu_-(z) \). Continuity of these functions at \( z = 0 \) and \( z = d \) gives the following four conditions.

\[
A_F = \tilde{A}_N + \tilde{B}_N, \quad \tilde{A}_Ne^{i\phi_i}\tilde{B}_Ne^{-i\phi_i} = \tilde{B}_Ne^{-i\phi_i}, \quad \tilde{C}_N = C_N = e_{j_{\text{app}}}.
\]

We now put these into the solution and obtain

\[
\mu_+(z) - \mu_-(z) = \begin{cases} (\tilde{A}_N + \tilde{B}_N)e^{i\phi_i} & \text{for } z < 0, \\ \tilde{A}_Ne^{i\phi_i} + \tilde{B}_Ne^{-i\phi_i} & \text{for } 0 < z < d, \\ (\tilde{A}_Ne^{i\phi_i} + \tilde{B}_Ne^{-i\phi_i})e^{-(z-d)l_p} & \text{for } z > d, \end{cases}
\]

\[
\sigma_+(z)\mu_+(z) + \sigma_-(z)\mu_-(z) = \begin{cases} e_{j_{\text{app}}}z + D_F & \text{for } z < 0, \\ e_{j_{\text{app}}}z & \text{for } 0 < z < d, \\ e_{j_{\text{app}}}z + D_N & \text{for } z > d. \end{cases}
\]

Now we apply the continuity of \( \mu_+(z) + \mu_-(z) \). After some algebra,

\[
\mu_+(z) + \mu_-(z) = \begin{cases} \frac{2e_{j_{\text{app}}} + 2D_F}{\sigma_+ + \sigma_-} & \text{for } z < 0, \\ \frac{e_{j_{\text{app}}}z}{\sigma_-} & \text{for } 0 < z < d, \\ \frac{e_{j_{\text{app}}}z + D_N}{\sigma_-} & \text{for } z > d. \end{cases}
\]

Continuity at \( z = 0 \) and \( z = d \) gives

\[
D_F = \frac{\sigma_+ - \sigma_-}{2}(\tilde{A}_N + \tilde{B}_N), \quad D_N = \left( \frac{\sigma_-}{\sigma_-} - 1 \right) e_{j_{\text{app}}}d.
\]

Then \( \tilde{A}_N \) and \( \tilde{B}_N \) are the only remaining coefficients. We now apply continuity of \( j_+(z) - j_-(z) \). After some algebra,

\[
\sigma_+(z)\mu_+(z) - \sigma_-(z)\mu_-(z) = \begin{cases} \frac{\sigma_+ - \sigma_-}{\sigma_+ + \sigma_-}e_{j_{\text{app}}}D_F + \frac{2\sigma_+\sigma_-}{\sigma_+ + \sigma_-}(\tilde{A}_N + \tilde{B}_N)e^{i\phi_i} & \text{for } z < 0, \\ \sigma_-\tilde{A}_Ne^{i\phi_i} + \tilde{B}_Ne^{-i\phi_i} & \text{for } 0 < z < d, \\ \sigma_-\tilde{A}_Ne^{i\phi_i} + \tilde{B}_Ne^{-i\phi_i})e^{-(z-d)l_p} & \text{for } z > d. \end{cases}
\]

Continuity of the derivatives at \( z = 0 \) and \( z = d \) gives

\[
\frac{\sigma_+ - \sigma_-}{\sigma_+ + \sigma_-}e_{j_{\text{app}}} + \frac{2\sigma_+\sigma_-}{\sigma_+ + \sigma_-}\frac{\tilde{A}_N + \tilde{B}_N}{l_p} = \frac{\sigma_-}{l_p}(\tilde{A}_N - \tilde{B}_N).
\]

\[
\frac{\sigma_-}{l_p}(\tilde{A}_Ne^{i\phi_i} - \tilde{B}_Ne^{-i\phi_i}) = \sigma_-\tilde{A}_Ne^{i\phi_i} + \tilde{B}_Ne^{-i\phi_i),
\]

the solutions of which are

\[
\tilde{A}_N = e^{-2ld_i}\tilde{A}_N\tilde{N} + \tilde{B}_N, \quad \sigma_- \tilde{N} \tilde{N} + \tilde{B}_N,
\]

\[
\tilde{B}_N = \frac{\sigma_+ - \sigma_-}{\sigma_+ + \sigma_-}e_{j_{\text{app}}} + \frac{2\sigma_+\sigma_-}{\sigma_+ + \sigma_-}e^{-2ld_i}\tilde{A}_N - \tilde{B}_N\tilde{N} - \sigma_-\tilde{A}_N\tilde{N} + \tilde{B}_N(e^{-2ld_i}\tilde{B}_N - \tilde{A}_N) \sigma_-\tilde{A}_N\tilde{N} - \sigma_-\tilde{A}_N\tilde{N} + \tilde{B}_N(e^{-2ld_i}\tilde{B}_N - \tilde{A}_N) \sigma_-\tilde{A}_N\tilde{N}.
\]
$D_F$ and $D_N$ are determined by Eq. (A9). Thus, we determine all coefficients of Eqs. (A7) and (A6).

**Appendix B: Scaling procedure of the experimental data**

Due to inevitable sample-to-sample variations, and potential changes in interface resistance, limited information can be extracted from changes in the absolute magnitude of $\alpha_{\text{eff}}$ on annealing. This however does not preclude an analysis of the changes to Kondo depolarization, provided a method is established to appropriately scale $\alpha_{\text{eff}}(T)$. In this section we will outline the procedure applied to reach the scaled data of Fig. 3(a).

The Kondo expression of Eq. (10) is valid only over a narrow range about $T \approx T_K$, and one of the major successes of the G-G formalism was to accurately describe the evolution of $\rho_K(T)$ from low ($T \ll T_K$) to high $T$ ($T \gg T_K$). It is worth noting at this point the limiting values of $\rho_K$ in these two regimes. At low $T$ the Kondo effect saturates towards a constant scattering rate as the unitary limit is reached, which Kondo proposed to give $\rho_K \rightarrow \rho_m [1 + 2N_0 J_{\text{in}} (U/T^*)]$. At high $T$ the effect is negligible and $\rho_K$ tends to the classical constant expression for spin-flip scattering via exchange with the ferromagnetic impurity $\rho_K \rightarrow \rho_m$ (i.e. the Korringa rate).

Although we cannot a priori determine the magnitude of these two regimes for our experimental data, by considering the data of reference [31] and the transition temperatures between the three regimes we can deduce $\rho_K(T \ll T_K)/\rho_K(T \gg T_K) = 1 + 2N_0 [\ln (U/T^*)] \approx 1.8$.

Using Eq. (9) we may obtain an experimental estimate of $\rho_K(T)$ at each $T_A$, which is explicitly dependent upon $\alpha_{\text{eff}}(T)$.

It is worth noting that $\rho_N$, $P_F$, $t_{ff}^T$, $t_{ff}^N$ and $\alpha_{\text{FM}}$ are measured directly, while $\eta$, $d$ and $r_N^{ff}/\tau_N$ are all $T$-independent, leaving only the scaled value of $\alpha_{\text{eff}}$ undetermined in Eq. (9). To appropriately normalize $\alpha_{\text{eff}}(T)$ to $\alpha_{\text{FM}}(T)$ we linearly scale $\alpha_{\text{eff}}(T)$ (and consequently modify $\rho_K(T)$) in order to reach the correct ratio of $\rho_K$ at low- and high-$T$ (i.e. $\rho_K(T \ll T_K)/\rho_K(T \gg T_K) = 1.8$). Note, that since scaling $\alpha_{\text{eff}}$ in this way only ensures the correct ratio of Kondo to classical scattering, we may still fit $1 - k$ to obtain the magnitude of the scattering (both Kondo and classical) and therefore deduce $\eta$.

Once we have established correct normalization for a single dataset (in this case the unannealed data), we may normalize the remaining data by observing the following relationship from Eq.10 and Eq.11:

$$\frac{\rho_K'(T)d^i}{\rho_K'(T)d^j} = \frac{d^i C_{Fe}'}{d^j C_{Fe}'} = \text{const.}$$  \hspace{1cm} (B1)

Where, the superscript $i$ denotes values for different $T_A$. Note this relationship exploits the fact that annealing only serves to increase the magnitude of $d$ and $C_{Fe}$ in $\rho_K$, both of which are $T$ independent, thus the functional form of $\rho_K(T)$ is independent of $T_A$. From Eq. (9):

$$\rho_K'(T)d^i = (1 - r_i') \left[ \left( \frac{2\eta \tau_{ff}^N/\tau_N}{\rho_N} \right) \left( \frac{1}{\lambda_N^i} - \frac{1}{\lambda_N + \frac{\lambda_{ff}^i}{(1 - \alpha_{ff}^i)}} \right) \right]^{-1}.$$  \hspace{1cm} (B2)

Here $r'$ is the scaling factor for $\alpha_{\text{eff}}(T)$. Since the ratio of $\rho_Kd$ is constant, we minimize the standard deviation of expression B1 by varying $r'$, to ensure $\alpha_{\text{eff}}(T)$ is correctly scaled at each $T_A$.

**Appendix C: Mathematical details for the derivation**

1. **Legendre decomposition of the Boltzmann equation**

We first expand the first term in Eq. (12) by the Legendre polynomial. The Bonnet recursion formula is useful to do this.

$$xP_n(x) = \frac{n + 1}{2n + 1} P_{n+1}(x) + \frac{n}{2n + 1} P_{n-1}(x),$$ \hspace{1cm} (C1)

for $n \geq 1$.

$$\frac{\hbar k}{m} \partial_z g_s(z, k) = \frac{\hbar k}{m} \sum_{n=0}^{\infty} \partial_z g_{n}^{(m)}(z, k) \cos \theta_k P_n(\cos \theta_k)$$

$$= \frac{\hbar k}{m} \partial_z g_s^{(0)}(z, k) P_1(\cos \theta_k) + \frac{\hbar k}{m} \sum_{n=1}^{\infty} \partial_z g_{n}^{(m)}(z, k) \left[ \frac{n + 1}{2n + 1} P_{n+1}(\cos \theta_k) + \frac{n}{2n + 1} P_{n-1}(\cos \theta_k) \right]$$

$$= \frac{\hbar k}{m} \sum_{n=0}^{\infty} \left[ \frac{n}{2n - 1} \partial_z g_{n-1}^{(m)}(z, k) + \frac{n + 1}{2n + 3} \partial_z g_{n+1}^{(m)}(z, k) \right] P_n(\cos \theta_k).$$ \hspace{1cm} (C2)

The second term in Eq. (12) is

$$-\frac{eE}{\hbar} \partial_z f_{s_0}^{(k)} = -\frac{eE}{\hbar} \partial_k f_{s_0}^{(k)} \times P_1(\cos \theta_k).$$ \hspace{1cm} (C3)
The right-hand side of Eq. (12) is
\[-g_s(z,k) - g_s(0,z,k) \tau^T(z,k) = -g_s^{(0)}(z,k) - g_s(0,z,k) \tau^T(z,k) - \frac{1}{\tau_s(z,k)} \sum_{n=1}^{\infty} g_s^{(n)} P_n(\cos \theta_k), \tag{C4}\]
where $1/\tau_s = 1/\tau^T_s + 1/\tau^F_s$.

In summary, Eq. (12) is equivalent to
\[\sum_{n=0}^{\infty} \left[ \frac{\hbar k}{m} n \frac{\partial}{\partial n} g_s^{(n-1)}(z,k) + \frac{\hbar k}{m} n + 3 \frac{\partial}{\partial n} g_s^{(n+1)}(z,k) = \frac{eE}{\hbar} \delta f_s^0(k) \delta \delta_{n,1} \right] P_n(\cos \theta_k) \]
\[= -g_s^{(0)}(z,k) - g_s(0,z,k) \tau^T(z,k) - \frac{1}{\tau_s(z,k)} \sum_{n=1}^{\infty} g_s^{(n)} P_n(\cos \theta_k). \tag{C5}\]

Since \(\{P_n\}\) forms an orthogonal set of polynomials, each coefficient should satisfy the equation. In Ref. [17], if the spin diffusion length is much larger than the mean free path of conduction electrons, \(g_s^{(2)}\) (and higher order terms) can be neglected. The coefficients of \(P_0\) and \(P_1\) gives Eqs. (13) and (14), once \(g_s^{(2)}\) is neglected.

2. Sommerfeld expansion formula

In this section, we present the Sommerfeld formula for low temperature. In the main text, we keep terms up to \(O(T)\), we here present the formula up to \(O(T^3)\) for more motivated readers.

The Sommerfeld expansion formula for a differentiable function \(H\) is
\[\int \frac{H(\epsilon)}{e^{\beta(\epsilon-\mu)} + 1} d\epsilon = \int H(\epsilon) d\epsilon + \frac{\pi^2}{6\beta^2} H'/(\mu) + O(T^4). \tag{C6}\]
In a compact form, as far as quantities after integration over \(\epsilon\) is concerned,
\[\frac{1}{e^{\beta(\epsilon-\mu)} + 1} = \Theta(\mu - \epsilon) + \frac{\pi^2}{6\beta^2} \delta(\mu - \epsilon) + O(T^4). \tag{C7}\]
When a transport property is concerned, it is convenient to take the derivative with respect to \(\epsilon\).
\[\frac{\partial}{\partial \epsilon} \frac{1}{e^{\beta(\epsilon-\mu)} + 1} = -\delta(\mu - \epsilon) - \frac{\pi^2}{6\beta^2} \delta'^{'}(\mu - \epsilon) + O(T^4). \tag{C8}\]

3. Integrals including the Kondo scattering rate

In this section, we perform the following integration for a general \(G(\epsilon)\)
\[\int d\epsilon G(\epsilon) \gamma(\epsilon) \partial f_0, \tag{C9}\]
where \(f'^0 = [1 + e^{\beta(\epsilon-\mu)}]^{-1}\) and \(\gamma(\epsilon)\) is defined by Eq. (24). Here and from now on, we denote \(k = \sqrt{2m\epsilon/h}, k' = \sqrt{2m\epsilon'/h}\) and so on, appearing in integrations with respect to \(\epsilon\) and \(\epsilon'\). Also, we define \(k^2 = \sqrt{2m\mu h}/h\) which is the Fermi wave vector. We generalize the approach taken by Kondo [31] here.

First we perform the summation in \(\gamma(\epsilon)\).
\[\gamma(\epsilon) = \frac{1}{V} \sum_k \frac{f'^0(k)}{\epsilon(k)-\epsilon} = \frac{1}{8\pi^3} \int \frac{d^3k'}{e(k')-\epsilon} = \frac{1}{2\pi^2} \int \frac{d\epsilon' k'^2}{e(k') - \epsilon} \]
\[= \frac{m}{\pi^2 \hbar^2} \int d\epsilon' \frac{e(k')}{e(k') - \epsilon} f'^0(k') = \frac{m}{\pi^2 \hbar^2} \int d\epsilon' f'^0(k') + \frac{m\epsilon}{\pi^2 \hbar^2} \int d\epsilon' \frac{f'^0(k')}{e(k') - \epsilon}. \tag{C10}\]
The first integral can be given by the Sommerfeld expansion Eq. (C7).
\[\frac{m}{\pi^2 \hbar^2} \int d\epsilon' f'^0(k') = \frac{m}{\sqrt{2\epsilon}} \int d\epsilon' \frac{1}{\sqrt{2\epsilon'}} f'^0(\epsilon') = 2N_0. \tag{C11}\]
To perform the second integral,
\[
\frac{me}{\hbar^2} \int dk' \frac{f^0(k')}{g(k') - \varepsilon} = \frac{2m^2 e}{\hbar^2} \int dk' \frac{f^0(k')}{k^2 - k'^2} = -\frac{m^2 e}{\hbar^2 k^4} \int dk' \ln \frac{k - k'}{k + k'} \cdot \partial_{k'} f^0. \tag{C12}
\]
We are now ready to perform the integral in Eq. (C9).
\[
\int d\varepsilon g(\varepsilon) \partial_{\varepsilon} f^0 = 2N_0 \int d\varepsilon g(\varepsilon) \partial_{\varepsilon} f^0 - \frac{m}{2\pi^2 \hbar^3} \int d\varepsilon G(\varepsilon) \ln \frac{\varepsilon - \varepsilon'}{\varepsilon + \varepsilon'} \cdot \partial_{\varepsilon} f^0 \partial_{\varepsilon} f^0.
\]
\[
= 2N_0 \int d\varepsilon G(\varepsilon) \partial_{\varepsilon} f^0 - \frac{m}{2\pi^2 \hbar^3} \int d\varepsilon G(\varepsilon) \ln \frac{\varepsilon - \varepsilon'}{\varepsilon + \varepsilon'} \cdot \partial_{\varepsilon} f^0 \partial_{\varepsilon} f^0. \tag{C13}
\]
The first integral is given by the Sommerfeld expansion Eq. (C8). For the second term, exact substitution of $X = \beta(\varepsilon - \mu^0)$ and $X' = \beta(\varepsilon' - \mu^0)$, the second term is
\[
-\frac{m}{2\pi^2 \hbar^3} \int d\varepsilon G(\varepsilon) \ln \frac{\varepsilon - \varepsilon'}{\varepsilon + \varepsilon'} \cdot \partial_{\varepsilon} f^0 \partial_{\varepsilon} f^0 \approx -\frac{m}{2\pi^2 \hbar^3} k^2 G(\mu^0) \ln \frac{k T}{\mu^0}.
\tag{C14}
\]
Here we expanded with respect to $k T$, which is small compared to $\mu^0$. We drop the second contribution $\ln(X - X')/4$ since it gives a much smaller contribution than the $\ln k T / \mu^0$ contribution at low temperature. Thus we keep only the logarithmic term.
\[
\int d\varepsilon G(\varepsilon) \ln \frac{\varepsilon - \varepsilon'}{\varepsilon + \varepsilon'} \cdot \partial_{\varepsilon} f^0 \partial_{\varepsilon} f^0 \approx -\frac{m}{2\pi^2 \hbar^3} k^2 G(\mu^0) \ln \frac{k T}{\mu^0}. \tag{C15}
\]
By using $N_0 = mk^2 / 2\pi^2 \hbar^2$, for low temperature, the following replacement is valid under an energy integration.
\[
\gamma(\varepsilon) \partial_{\varepsilon} f^0 \to -N_0 \ln \frac{k T}{\mu^0} \delta(\varepsilon - \mu^0), \tag{C16}
\]
which is Eq. (26).

**Appendix D: Error analysis**

Errors in the parameters $p_N^0$, $\alpha_{\text{eff}}$, $\rho_N$, $\rho_F$ (experimentally determined) and $\kappa$ (determined through a normalization procedure), as well as uncertainty from our fitting method are our main concern in establishing uncertainty in the extracted values of $\eta$. All other parameters are constrained through the previous work, and potential errors in such quantities are not considered.

Although both $\rho_N$ and $\rho_F$ are measured directly from $R_N$ and $R_F$, with very small random noise, these quantities suffer from experimental uncertainty in the wire cross sectional area (through $\rho = R A / L$), particularly a non-rectangular shape. This uncertainty is random between devices, but systematic across all $T$ within a single device. It is estimated at the level of $\pm 5\%$ from SEM images of the wire edge profile. In our measurement of $\Delta R_{\text{NL}}$, we observe a baseline noise floor of around 1 nV (at a modulation frequency of 13 Hz). This corresponds to $\approx 3 \mu\Omega$ in our measurements and is an absolute noise source independent of signal size. When fitting $\Delta R_{\text{NL}}(L)$ at each temperature to extract $p_N^0$ and $\alpha_{\text{eff}}$, the uncertainties in $\rho$ and $\Delta R_{\text{NL}}$ are used to weight a least means square minimization fit, with estimated errors in $p_N^0$ and $\alpha_{\text{eff}}$ arising from combining these errors with the fitting residuals. In reality, the parameters $p_N^0$ and $\alpha_{\text{eff}}$, are limited in precision by the relative uncertainty in the cross-sectional area measurements, and are therefore largely independent of $T$. Although we may obtain $\alpha_{\text{eff}}$ and $p_N^0$ by fitting $\Delta R_{\text{NL}}(L, T)$, using a literature value of $p_N^0$ as a constraint on the signal magnitude, the magnitude of $\alpha_{\text{eff}}$ is poorly constrained, due to the inherent difficulty in precisely measuring the ferromagnet/normal metal interface resistance. Consequently, errors that are independent of $T$ dominate the extracted values of $\alpha_{\text{eff}}$.

With the errors for $p_N^0$ and $\alpha_{\text{eff}}$ established, it remains to estimate the uncertainty in $\kappa$, before determining $\eta$. As both $\alpha_{\text{FM}}$ and $\alpha_{\text{eff}}$ are broadly of a similar magnitude, and $\kappa = \alpha_{\text{eff}} / \alpha_{\text{FM}}$, the systematic errors in each quantity could, in principle, give an error larger than the estimated value of $1 - \kappa$ (typically $1 - \kappa$...
is around 10%, while errors in $\alpha$ are around 5% to 10%. However, this systematic error is irrelevant for the normalization procedure we use, and is one of the key advantages to our method: As any error in $\alpha$ are largely T-independent (errors from both fitting and estimates of interface resistance), they make no impact on the overall normalization factor [$r$ in Eq. (B2)], since any systematic error in $\alpha_{\text{fit}}$ or $\alpha_{\text{FM}}$ is intrinsically compensated by $r$. Thus we can estimate the error in $\kappa$ solely from the uncertainty in the normalization procedure. To obtain this value, we realize that the process of minimizing the standard deviation of $\rho_{\kappa} f / \rho_{\kappa} d$ (our normalization procedure) is identical to a linear regression of $y = A(1 - r\kappa)$, where $y = \rho_{\kappa} f / \rho_{\kappa} [1/(\rho_{\kappa} f - 1)] + \rho_{\kappa} d \{(1 - \alpha_{\text{FM}}) \rho_{\text{F}} - 1\}]$, and $A = C_{\text{Fe}} d^2 \rho_{\text{Fe}}^{-1} / 2C_{\text{Fe}} d^2 \rho_{\text{Fe}}^{-1}$. Once again, we use the superscript $i$ to denotes a given dataset (i.e. a given $T_A$), with $i = 1$ representing the unannealed data (which can be scaled exactly, see Appendix B). As we can establish both $y$ and $\kappa$ for a given $T_A$, we may therefore estimate the uncertainty in $r$, and so the relative error in our scaled $\kappa$, from the residuals of a least squares fit of $y = A(1 - \kappa)$ with $A$ and $r$ as free variables. These estimates are the error bars shown in Fig 4.

The final challenge is to incorporate all these errors together for our final fitting procedure to estimate $\eta$. $\eta$ is found from fitting $\eta \rho_{\kappa}$ from experimental data using our models for $\rho_{\kappa}$, i.e. through rearranging Eq. (9). Through the discussed procedure we now have estimates for all parameters in this equation, including errors for the experimentally determined quantities ($\kappa, \rho_{\kappa}, \rho_{\text{F}}, \alpha, l_{\text{F}}$). To establish the error on the experimental $\eta \rho_{\kappa}$ we use Monte Carlo sampling assuming Gaussian distributed uncertainties for all quantities (via the NIST uncertainty machine [46]) to account for the combination of all uncertainties in Eq. (9). The extracted errors are subsequently used as weightings for fitting $\eta \rho_{\kappa}$ to either the G-G or Kondo model, again using a least-mean-square approach. The extracted parameter uncertainties are shown in Fig 4 for each method, with the final errors for $\eta$ in panel (c). All quoted errors are a single standard deviation, including those shown for the extracted values of $T_K$. Most errors are relative rather than absolute, and so data at large $T_A$ show more error than those at low $T_A$, despite the larger $\kappa$. In calculating $\bar{\eta}$ an unweighted average is taken, with the uncertainty in this case quoted as the standard deviation in the eight values.

[47] µmol/mol is equivalent to ‘parts per million’
[48] We emphasize that each region is specified by subscripts. Note that the superscript $F$ that appears in $\kappa_F^2$ for instance refers to at the Fermi surface.
[49] One can verify explicitly that $\kappa$ has the same value for both definitions.
[50] \[ \int_{C_{Fe}=2600\mu mol/mol}^{\infty} [1 - \text{erf}(x/L_{Fe})] dx = 560 \mu mol/mol \cdot L_{Fe} \]
[51] See Eq. (12) of Ref. [31].