Direct Experimental Evidence for the Ruderman-Kittel-Kasuya-Yosida Interaction in Rare-Earth Metals

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We show that the ferromagnetic heavy rare-earth (RE) metals show a transport spin polarization at the Fermi level in the majority spin, whereas in ferromagnetic light rare earths it is in the minority spin. The sign of the polarization is in agreement with what is expected due to the Ruderman-Kittel-Kasuya-Yosida (RKKY) coupling formalism. We show that magnetotransport measurements on magnetic multilayer samples containing magnetic REs provide a unique opportunity to verify the RKKY coupling scheme in pure rare-earth metals, allowing us to probe both the sign and temperature dependence of the spin-density oscillation.

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The origin of magnetic order in the Lanthanide metals has been a fundamentally interesting topic in magnetism for nearly half a century [1,2]. The theoretical explanation of the coupling between ionic $4f$ magnetic moments due to an exchange interaction mediated by the conduction electrons is now accepted in the scientific community. The model of magnetic coupling mediated by an oscillatory spin polarization of the conduction band has come to be known as the Ruderman-Kittel-Kasuya-Yosida (RKKY) indirect exchange interaction [1,3,4]. Despite the degree of acceptance that the RKKY theory has gained, the predicted spin polarization of the conduction band at the Fermi level [5] in the bulk of pure rare-earth (RE) metals has never in fact been directly verified by experiment in a systematic fashion to the best of the authors' knowledge. The magnetic polarization of the conduction band is known to contribute some fraction of the moment in the REs; in the case of Gd this is roughly 0.5 of a total moment of $7.5\mu_B$/Gd atom [6]. Previous investigations on the RKKY interaction have focused on obtaining the values of exchange integrals at, and coupling strengths between, various nuclear sites in magnetic alloy systems [2,7–9], metals [10], and bilayers [11]. We present in this Letter experimental evidence for the spin polarization of the conduction band due to the RKKY mechanism and show that the temperature dependence of the polarization is in sound qualitative agreement with the RKKY picture of the coupling interaction for all magnetic phases of the RE metals investigated.

Artificially layered magnetic structures provide an ideal means for investigating fundamental properties of magnetic materials [12]. The evidence for the RKKY spin polarization which we present is based on the giant magnetoresistance (GMR) [13,14], which arises due to spin-dependent scattering of conduction electrons in the ferromagnetic layers of ferromagnet/nonmagnetic multilayers. Transport measurements on GMR systems are intrinsically sensitive to the electronic spin polarization in each ferromagnetic layer [15], so by changing the constituent magnetic layers it is possible to determine the sign of the spin polarization in different materials.

In light of much of the recent work on spin-polarized transport, the question of what exactly is meant by "spin polarization" naturally arises [16]. For any transport process the popular definition of spin polarization, based simply upon the density of states at the Fermi level, is inapplicable due to the difference in mobility between the s, p, and d electron states. The conduction-band susceptibility, which gives the form of the RKKY range function $J(R)$, is dependent upon the density of states at E_F. The conduction electron spin polarization which actually carries the interaction is, however, that of the electric current

$$P = \frac{I_\uparrow - I_\downarrow}{I_\uparrow + I_\downarrow}.$$

In a simplistic case this reduces to the v_F^2 weighted polarization discussed in Ref. [16] which highlights the contribution to the transport properties due to the lighter sp electrons.

As it is the current-carrying electrons on the Fermi surface which effectively mediate the RKKY interaction, the spin polarization seen in these transport measurements is exactly that which is of interest in probing the magnetic ordering interaction in the rare earths.

In contrast to other recent work on the electronic and/or magnetic structures in the RE metals and their alloys, e.g., using spin-resolved photoemission [17] or x-ray resonant exchange scattering [18] which directly probe the whole density of states, our measurements are sensitive only to the spin polarization of the electronic levels which are directly implicated in mediating the bulk magnetic ordering interaction. Superconductor/ferromagnet tunneling [19] is sensitive mainly to the polarization of s-like electrons with quasimomenta predominantly perpendicular to the interface plane. The polarization probed is representative only of the region close to the interface [20], rather than the bulk of the material. The Andreev reflection technique [21] is unable to resolve the sign of the spin polarization. Both of these methods are also unable to probe higher temperature magnetic phases.

The multilayer samples reported in this Letter are deposited by dc magnetron sputtering in a custom vacuum chamber having a base pressure better than 2×10^{-8} Torr. Deposition rates are in the range 3–5 Å s^{-1}. The samples have multilayer structures of either $\{RE/Cu\}_N$ or $\{RE/Cu/Co/Cu\}_{N/2}$ where RE is one of Dy, Gd, or Nd, and N is the number of structural repeat units. Nominal layer thicknesses are confirmed by x-ray reflectivity, which yields an rms interfacial roughness of typically 5–6 Å, which is comparable with a monolayer thickness of the rare earth. Magnetoresistance (MR) measurements are performed in the current-in-plane (CIP) geometry and are carried out by the four-probe dc technique in a ^{4}He gas-flow cryostat. The field is applied in the plane of the sample, which can be rotated in the magnetic field. Magnetization measurements are made by variable temperature vibrating sample magnetometry (VSM).

We begin by discussing samples with structure $\{RE/Cu\}_N$. Magnetization measurements confirm that at our base measurement temperature, (8.23 ± 0.02) K, the rare-earth layers in all samples are ferromagnetically ordered. Figure 1 shows the MR of a $\{Dy[25 Å]/Cu[20 Å]\}_{60}$ multilayer, measured in transverse ($I \perp B$), diagonal and longitudinal ($I // B$) orientations and the corresponding angular dependence of high-field resistance, all measured at base temperature. The sample exhibits a small negative MR of about -0.5%, which agrees with the "less than 1%" previously reported in the REs [22]. Peaks appear symmetrically about zero applied field, implying that there is no strong interlayer coupling across the Cu spacer layers as is expected for samples such as these[23]. The data for Gd/Cu and Nd/Cu multilayers are, in this case, qualitatively similar.

Possible contributions to the MR we measure are due to the GMR, anisotropic magnetoresistance (AMR) [24], Lorentz MR [25], and the Kondo effect [26]. In order to show that the effects that we observe are attributable to the GMR we discuss briefly the AMR and Lorentz MR contributions.

We have performed MR measurements in the transverse, diagonal, and longitudinal orientations to show the contribution due to the AMR. The spin-orbit scattering of conduction electrons in a ferromagnet [24] manifests itself as a variation in the resistance as a function of the angle between magnetization and current. Figure 1(d) shows the variation of saturated resistance of the Dy/Cu multilayer with angle, the $\cos^2\theta$ dependence being typical of the AMR. In these samples the high-field resistance is always lower than that at zero applied field; the AMR accounts only for the difference in resistance at high field with angle, not the general form of the MR curves. We are thus able to separate the AMR and GMR in our measurements.

The Lorentz MR arises due to the action of the Lorentz force on the conduction electrons in a metal. It is generally several orders of magnitude smaller than that seen in our multilayers. The Lorentz MR is strongly material dependent, so varying the composition of the multilayer should produce a change in the negative MR of the sample if this is the dominant MR mechanism.

To show conclusively that the result presented above is due to spin-dependent scattering rather than the Lorentz MR, we discuss now samples where alternate

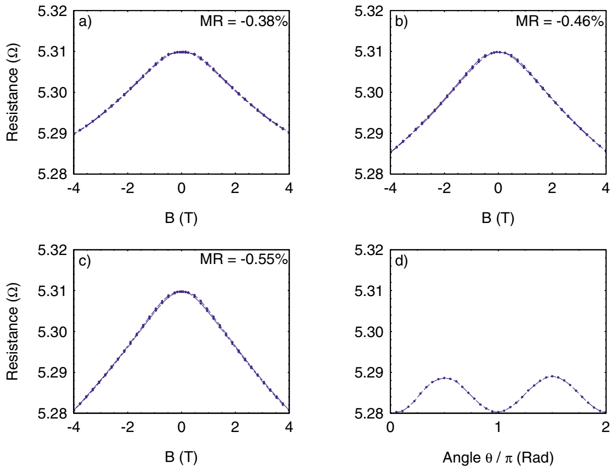

FIG. 1 (color online). MR of a Dy/Cu multilayer in (a) transverse, (b) diagonal, and (c) longitudinal orientations. Frame (d) shows the angular dependence of the resistance at 4 T. The 4 T resistance in frames (a)–(c) agrees with the resistances at $\theta = 0$, $\pi/4$, and $\pi/2$.

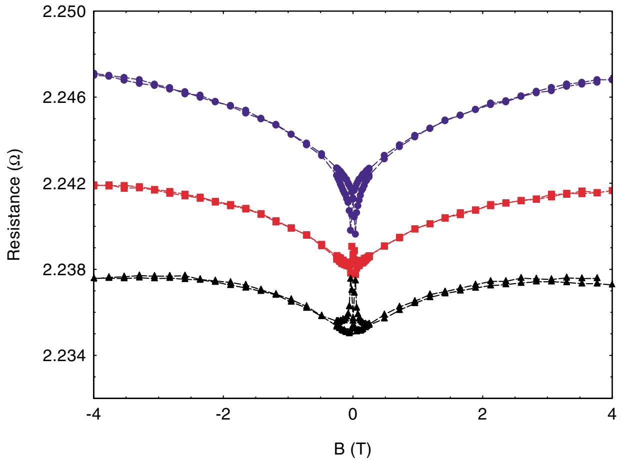

FIG. 2 (color online). MR of a Dy/Cu/Co/Cu multilayer in longitudinal (●, MR = 0.34%), diagonal (■, MR = 0.19%) and transverse (▲, MR = 0.12%) orientations. The overall shape of the GMR is now inverted with the sharp peaks close to zero field being the AMR in the Co layers.

Dy layers have been exchanged for an equal thickness of Co. Figure 2 shows the MR for a $\{\mathrm{Dy}[25\,\text{Å}]/\mathrm{Cu}[20\,\text{Å}]/\mathrm{Co}[25\,\text{Å}]/\mathrm{Cu}[20\,\text{Å}]\}_{30}$ multilayer. The shape of the MR curves has changed drastically in comparison to those in Fig. 1. The high-field resistance and sharp peaks close to zero applied field are consistent with the AMR of the multilayer stack and constituent Co layers, respectively. The same inversion is seen in our Gd/Cu and Gd/Cu/Co/Cu multilayers. The Lorentz MR in thin films is negative, independent of the material.

These results are evidence for spin-dependent scattering and therefore show that the conduction band of the RE is spin polarized. Polarization of the tunneling current in Dy and Gd has been observed previously [19], so it is not unreasonable to expect that a spin polarization may also be seen by conduction. As mentioned earlier, the polarization seen in tunneling experiments is not a probe of long-range magnetic order due to its localization close to the metal/insulator interface. The spin polarization of the conduction band explains the GMR observed in the Dy/Cu system, however, the question of why the GMR inverts upon exchanging alternate Dy layers for Co must now be answered.

There are three mechanisms reported in the literature whereby a positive GMR may occur in a multilayer structure. The first shows an inverse GMR only at low fields [27] and may be attributed to a magnetic spin-flop transition. Our samples do not exhibit strong interlayer coupling which is critical for a spin-flop transition to occur. The second is caused by direct antiferromagnetic coupling across a Dy/Co interface [28] which is not present in our samples. The final mechanism is due to the spin polarization of the conduction band having opposite sign in alternate layers [15]. Here the spin dependence of the scattering in alternate layers is reversed and the parallel magnetization state of the multilayer now has a higher resistance than the antiparallel magnetization state.

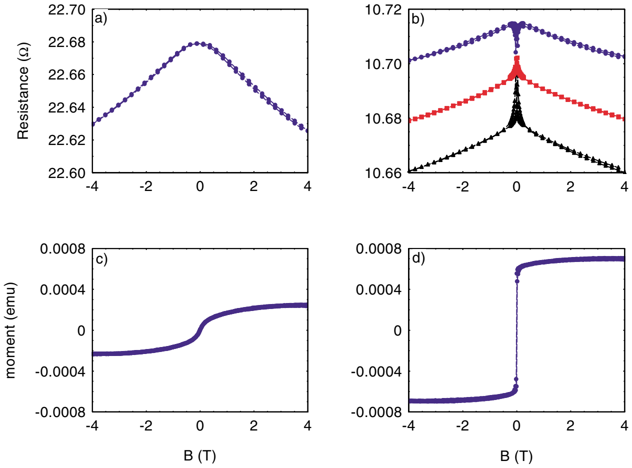

FIG. 3 (color online). MR of (a) a Nd/Cu multilayer in the longitudinal orientation and (b) a Nd/Cu/Co/Cu multilayer in longitudinal, diagonal, and transverse orientations. Symbols are as in Fig. 2. The corresponding longitudinal magnetization loops are shown in frames (c) and (d), respectively. Substituting alternate Nd layers with Co leaves the overall shape of the MR curves unchanged.

Assuming this final mechanism to be responsible for the inverse GMR in Dy/Cu/Co/Cu and knowing that the conduction spin polarization in Co is in the minority-spin channel, we may conclude that both of the heavy REs Dy and Gd have a net spin polarization in the majority spin.

It is instructive now to investigate the case of a light RE, such as Nd. Figure 3 shows the MR and magnetization curves for Nd/Cu and Nd/Cu/Co/Cu multilayer samples. In this case, apart from the expected resistance peaks due to the AMR in the Co layers, the curves do not change significantly. As the GMR in this multilayer remains negative we must conclude that the net spin polarization in ferromagnetic Nd is in the minority-spin direction, having an opposite sign to that in the heavy REs.

The change in sign of the spin polarization in crossing from light to heavy REs is direct evidence for the RKKY picture of interionic exchange coupling. The RKKY formalism predicts a net spin polarization [5] antiparallel to the spin S of the RE ion. Although both S and L are not necessarily "good" quantum numbers in the REs, the interaction between the spin-orbit angular momentum J of the ion and the conduction electron spin s has a prefactor $(g_J - 1)$, where g_J is the Landé factor, corresponding to a projection of S onto J [29]. Thus although the magnetic state of the ion is uniquely specified only by J, the spin polarization is always antiparallel to S. The projection of S along J, and hence along the ionic magnetic moment, can be either parallel or antiparallel, depending upon the spin-orbit coupling. In the light REs the ground state has J and S parallel, resulting in the induced

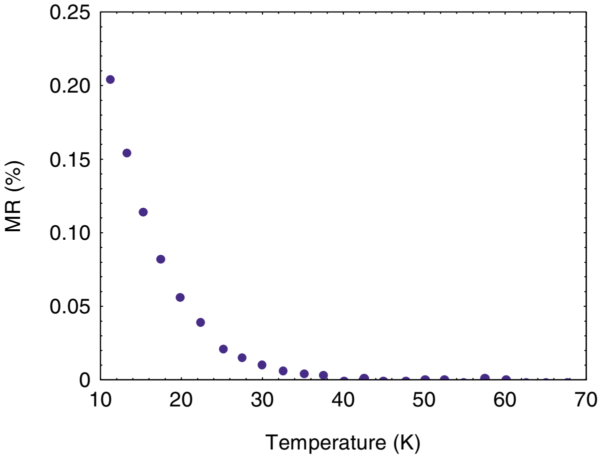

FIG. 4 (color online). Temperature dependence of the MR for a Dy/Cu multilayer in the longitudinal orientation.

spin polarization being antiparallel to J and hence in the minority spin. The heavy REs on the other hand have a ground state with S antiparallel to J, creating a spin polarization which is parallel to J so in the majority spin. Previously it has not been possible to measure the relationship between the ionic spin-orbit angular momentum J and the induced conduction-band spin polarization. It is noteworthy that any technique which probes the effective interionic coupling may at best measure the value of $(g_J - 1)^2$ [30] and thus gain no information on the spin polarization of the conduction band.

This argument is also applicable to the case of an antiferromagnetic (AF) RE; bulk Dy has a helical AF phase between 85 and 179 K [29]. In an AF or paramagnetic RE, the net spin polarization should vanish for either case of the spin-orbit coupling. Figure 4 shows the temperature dependence of the MR of a Dy/Cu multilayer. The MR tends to zero at around 40–50 K, remaining at this value for higher temperatures. The vanishing MR is due to the lack of spin polarization in the RE at these temperatures. AMR is observed for the sample containing Co, but no GMR is seen from the Co layers at these temperatures due to randomizing of the conduction electron spins upon crossing the RE layers.

To conclude, we have measured the sign of the spin polarization of the conduction electrons at the Fermi level in several RE multilayers. These measurements, and their temperature dependence, constitute the first direct experimental evidence for the RKKY theory of magnetic ordering in the REs. Although this theory is well accepted, it has not previously been possible to extract the polarization of the conduction electrons alone.

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