Anomalous behavior in the spin polarization of low-energy secondary electrons from Gd(0001)

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We have measured the spin-polarization spectrum of low-energy secondary electrons from the surface of thick single-crystalline Gd(0001) films grown on W(110). Below 1.5 eV an anomalous decrease in the polarization is observed and is attributed to the presence of the $4f^8$ states just above the vacuum level, which leads to increased emission of spin-down electrons. Between 1.5 and 7 eV the spin polarization remains nearly constant at about 33% (at 150 K), demonstrating that the phenomenon of polarization enhancement for low-energy secondary electrons is *not* universal. The observed large polarizations indicate that the completely polarized $4f$ electrons contribute significantly to the secondary-electron yield.

Spin-polarized secondary-electron emission at low kinetic energies has provided a powerful means in studying the magnetic properties of surfaces and ultrathin films.[1] On the other hand, study of the spin polarization of low-energy (<10 eV) secondary electrons may provide valuable information about the interaction between a hot electron and the ferromagnetic ground-state electrons and is therefore an interesting and important subject in its own right. Most of such studies to date have been performed on $3d$ transition-metal ferromagnetic materials, e.g., Ni, Fe, and Co.[2] Although the absolute magnitude of the spin polarization differs from one material to another, as might be expected from the rather different spin polarizations of their valence bands, which are the primary source for the generation of secondary electrons, their polarization versus energy spectra are strikingly similar. The most salient feature is the universal appearance of a large enhancement, by as much as a factor of 2–3, in the spin polarization of low-energy secondary electrons over the net valence-band polarization. With increasing kinetic energy, the polarization drops rapidly and approaches that of the valence band over an energy range of 10–20 eV.

The general picture that has emerged to explain such polarization behavior can be described by a three-step process, similar to the familiar three-step model often used in photoemission: excitation, transport to the surface, and escape into vacuum. In the first step, electrons are excited from the valence band to energies above the vacuum level, a process assumed to be spin independent. In a steady state this leads to a distribution of hot electrons in the solid with a spin polarization equal to the valence-band polarization. In the second step, these low-energy excited electrons undergo additional scattering processes which are spin dependent.[3,4] Inelastic scattering, including both spin-flip and nonflip processes, leads to preferential scattering of spin-down electrons due to the existence of only or predominantly spin-down empty states in the valence band, a common characteristic in the electronic structure shared by Ni, Fe, and Co.[5] The inelastic scattering acts as a spin filter, giving spin-up electrons a higher probability of reaching the surface and subsequently escaping into vacuum. At low energies, this spin filter effect reaches maximum[3,5,6] and causes significant enhancement in the spin polarization. The third step, escape into vacuum, can also show spin dependences and leads to structures in the polarization spectrum due to the elastic scattering.[7,8] However, these effects are nonuniversal and also smaller than the inelastic effects.

In this paper, we report the observation of anomalous behavior in the spin polarization of low-energy secondary electrons from a rare-earth ferromagnetic system: Gd. In contrast to the itinerant band ferromagnetism in $3d$ transition metals, Gd is a localized ferromagnetic system, with the bulk of its magnetic moment, $7\mu_B$ out of $7.63\mu_B$ per atom, carried by its seven localized $4f$ electrons occupying an energy level ($4f^7$) well below the Fermi energy ($E_B=8.3$ eV). Its valence band contains just three electrons, $(6s5d)^3$, and contributes $0.63\mu_B$ to the total atomic moment.[9] The low occupancy of the Gd conduction band ($\frac{3}{12}$) coupled with its moderate polarization (21%) leaves nearly as many spin-up as spin-down unfilled states above E_F. These important differences in the electronic structure between Gd and its $3d$ transition-metal counterparts suggest that the previous model on spin-polarized secondary-electron emission may not be simply carried over to describe Gd. Our experimental results indeed demonstrate not only that the universal polarization enhancement for low-energy secondary electrons is absent in Gd, but also that the spin polarization remains considerably larger than that of its valence band at higher energies. Most interestingly, the observation of an anomaly in the spin polarization below 1.5 eV indicates the existence of an additional channel for the emission of minority spin electrons.

The experiments were performed in a UHV system built for angle-resolved spin-polarized electron spectroscopies described elsewhere.[1] Gd films were evaporated onto a single-crystal W(110) substrate at a rate of 0.7–1.5 Å/sec from an e^--beam heated W crucible which is surrounded with a water-cooled shroud in a growth chamber equipped with a cylindrical mirror analyzer for Auger electron spectroscopy (AES) and a low-energy electron diffraction (LEED) optics for structural characteristics. After extensive outgassing of the Gd cell the base pressure during evaporation remained below 10^{-9} Torr and clean Gd films were obtained as verified with AES which showed only trace amounts of oxygen contamination [O_{KLL}(503 eV)/Gd_{CVV}(139 eV) <0.05]. Immediately after preparation the sample was cooled down to 150 K and magnetized with a magnetic field of a few hundred Oersted applied in the hcp(0001) film plane about 15° from one of the three symmetry axes. Secondary electrons were excited by a 1-keV electron beam impinging on the surface at 50° off-normal and collected around normal emission angles with the sample biased at -30 V with respect to the ground in order to suppress stray electrons. Energy analysis was achieved by the use of a 90° analyzer to which two orthogonally mounted medium-energy (20–30 kV) retarding-field Mott polarimeters (calibrated to within 10%) were coupled, allowing for the determination of all three components of the polarization vector. All polarization measurements were performed on samples in their remanent state. The overall energy resolution was about 400 meV.

The Gd films that we studied include two types of thick films (400–450 Å) that were deposited under different growth conditions. One type was grown at a substrate temperature of 400–450 °C, and the other type was grown at room temperature and subsequently annealed to 550 °C for 3–5 min. While growth at elevated substrate temperatures achieved good epitaxy, room-temperature growth resulted in disordered films as judged from diffuse LEED patterns. Annealing to >400 °C restored the ordered hcp(0001) structure as revealed by sharp 1×1 LEED patterns of comparable quality to that of films grown at elevated substrate temperatures, in agreement with earlier reports.[10,11] *In situ* magneto-optical Kerr effect (MOKE) measurements indicated that both types of films are 100% remanent at 150 K.[12] The polarization of secondary electrons obtained from the room-temperature grown films are found to be higher than that from films grown at 400–450 °C by a factor of about 1.4. Other than the difference in the absolute magnitudes of spin polarizations, however, both the polarization and intensity spectra for the two types of films are virtually identical. The higher polarization measured from films grown at room temperature is presumably due to reduced surface roughness, since Gd grows in a layer-by-layer Frank-van der Merwe mode at room temperature as opposed to three-dimensional Stranski-Krastanov (SK) growth at high temperatures.[13] In order to focus on the emphasis of the present study, we will restrict our discussion only on films grown at room temperature.

Figure 1 shows the intensity and polarization spectra of the low-energy secondary electrons between 0–7 eV

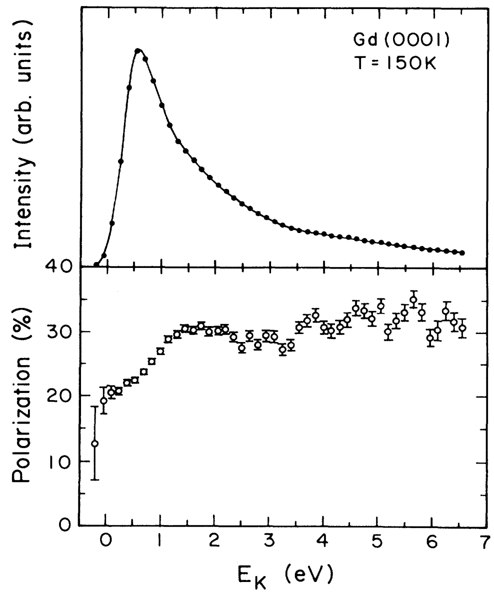

FIG. 1. Intensity spectrum (upper panel) and polarization spectrum (lower panel) of secondary electrons from Gd(0001).

kinetic energy measured at 150 K. The difference of the polarization spectrum from those of 3*d* transition-metal ferromagnetic surfaces is striking. Not only is there no significant enhancement at low energies but, on the contrary, the polarization shows a marked drop below 1.5 eV. Such drastically different behavior is closely related to the different electronic structure of Gd. Since in Gd the unoccupied valence-band density of states for spin-up electrons is only slightly smaller than for spin-down electrons, the inelastic mean free path for excited electrons should be nearly spin independent. Thus, the mechanism that leads to the polarization enhancement at low energies in 3*d* transition metals, i.e., the spin filter effect during transport to the surface is effectively suppressed in Gd. This result demonstrates that the phenomenon of polarization enhancement for low-energy secondary electrons is not universal.

The previous consideration, however, fails to account for the observed polarization drop below 1.5 eV. We attribute this decrease to the presence of the empty spin-down $4f^8$ states that lie about 1 eV above the vacuum level.[14] This state consists of seven closely spaced levels (due to spin-orbit interaction) and has a total width of ~1 eV.[14] The mechanism that we propose to account for the observed polarization drop is as follows. Excited electrons are scattered into empty states. In addition to the available conduction-band states, spin-down hot electrons can also scatter into the quasibound $4f^8$ states. These electrons may subsequently be emitted into vacuum via quasielastic one-electron decay processes with their minority spin character preserved. The energetics of the relevant electronic states are shown schematically in Fig. 2. Since this process only selectively enhances the emission of spin-down secondary electrons, the overall spin polarization will be reduced for energies

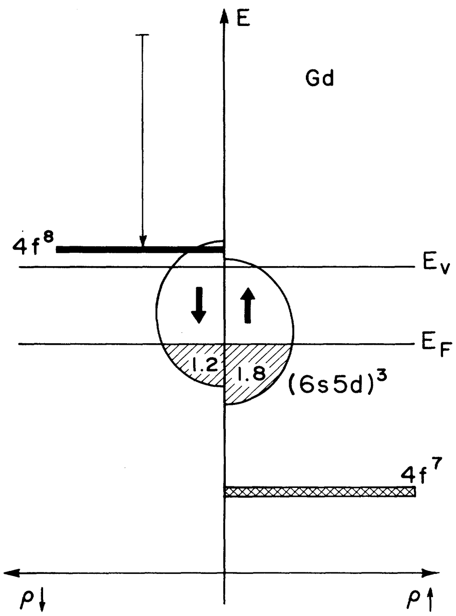

FIG. 2. Schematic of the electronic structure of Gd. The arrow indicates a scattering channel for hot spin-down electrons with the $4f^8$ level as an intermediate state which ultimately leads to emission into vacuum.

below ~ 1.5 eV. The upper bound 1.5 eV is estimated from the fact that the $4f^8$ states lie about 1 eV above the vacuum level and have a half-width of about 0.5 eV. Therefore, a drop in the spin polarization should be expected below 1.5 eV, in excellent agreement with our experimental result. We should mention that this process was proposed by Mauri and Landolt[15] to explain a structure in the polarization behavior in spin-polarized photoemission but was later disputed by Reihl and Himpsel.[16] Regardless of this controversy, Mauri and Landolt's discussions on how the one-electron decay process from a $4f^8$ state into a vacuum state may occur remain valid and are relevant to the present study.

In order to discuss the absolute magnitude of the spin polarizations, we must first consider the complicated nature of the surface magnetic state of Gd(0001). Unlike the simple ferromagnetic order found, e.g., on $3d$ transition-metal surfaces, Gd(0001) shows surface-enhanced magnetic order (SEMO) with a surface critical temperature well above the bulk value.[17–19] As the present study is concerned with the low-temperature (150 K) magnetizations only, however, SEMO does not play an important role. Further complication comes from the postulated antiferromagnetically coupled surface layer as the magnetic ground state of Gd(0001),[17,20] which would, of course, drastically affect the measured spin polarization owing to the high surface sensitivity of secondary electrons. While we find indeed that the surface magnetic state of Gd(0001) may be rather complicated,[19] on the films used in the present study we find no evidence of an antiferromagnetic surface layer. This is based on spin-polarized $4f$ photoemission measurements which show very large polarizations, 66% at 150 K. The results of this study will be published elsewhere.[19] Here it suffices to say that by extrapolating the temperature-dependent $4f$ polarizations one arrives at essentially complete polarization, i.e., 100%, at $T=0$ (within the accuracy of the extrapolation and the calibration of the spin detector), indicating that the surface is ferromagnetically coupled to the bulk. Thus, since we already know that there is no significant polarization enhancement mechanism at work in Gd, the measured secondary-electron polarization can be simply identified with the true polarization of the excited electrons upon irradiation with the primary beam.

Figure 1 shows that in the energy range of 1.5 to 7 eV the spin polarization of secondary electrons is almost constant at about 33%. Measurements at higher kinetic energies up to 15 eV revealed that the polarization continues to show its nearly constant behavior. Therefore, the value of 33% can be regarded as truly representing the polarization of the electrons in Gd that contribute to the secondary-electron yield at 150 K. To take into account the finite-temperature effects, we measured the representative spin polarization of 2 eV secondary electrons over a temperature range from 140 to 230 K and extrapolated the polarization to $T=0$ K. The data, shown in Fig. 3, yield a zero-temperature polarization of 43% with a $T^{3/2}$ fit and over 50% with a linear fit, which is only slightly inferior to the $T^{3/2}$ fit. Thus, a good estimate for the zero-temperature polarization is about 45%, which is much greater than the expected 21% valence-band polarization in Gd. This discrepancy firmly establishes that, in addition to the conduction electrons, the localized $4f$ electrons which are 100% polarized at 0 K also contribute significantly to the secondary-electron yield. The fact that the $4f^7$ state is only about 8.3 eV below the Fermi level lends credibility to the conclusion that this state is active in generating secondary electrons. As the polarizations for the $4f$ and conduction bands are both known, quantitative information about the $4f$ electron contribution may then be extrapolated from the polarization data. Based on a simple weighted average, the zero-temperature polarization can be expressed as

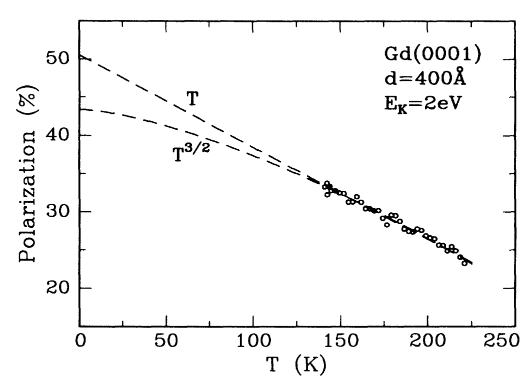

FIG. 3. Temperature dependence of the spin polarization of 2-eV secondary electrons.

$$P_0 = R + 0.21(1-R) , \quad (1)$$

where R is the normalized $4f$ electron contribution to the secondary-electron yield. For $P_0 = 0.45$, as found in the present study, we have $R = 0.3$. In other words, the $4f$ electrons contribute almost half as much as the conduction band does. It can be further inferred that on the average the ratio of the cross sections of a $4f$ electron and a conduction electron being excited upon irradiation with 1-keV primary electrons is about 0.18, which is rather significant. We note that Paul, Toscano, and Landolt[21] have previously measured the spin polarization of secondary electrons from polycrystalline Gd films grown on Ni and found the polarization to be larger than that of the valence band. However, they did not measure close enough to the vacuum threshold to see the drop in polarization.

In conclusion we have shown that the low-energy secondary electrons from epitaxial Gd(0001) films are highly polarized with significant contributions from the localized $4f$ electrons as well as from the valence bands. This finding opens the field of spin-polarized secondary-electron spectroscopy to rare-earth systems. We find no indication of a significant polarization enhancement, as seen in the $3d$ transition metals. Below 1.5 eV the polarization drops markedly. This anomaly is attributed to the presence of the $4f^8$ states which provide an additional scattering channel for spin-down electrons and subsequently leads to emission into vacuum.

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