The composite Hall effect of non-magnetic and magnetic bilayers

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Abstract

We verify, in Mn:Zn bilayers, that the Hall conductivity is averaged in a bilayer of non-magnetic or ferro- and paramagnetic materials, as previously reported, with thickness as a weighting factor, and also verify that this can lead to anomalously large Hall coefficients. We extend these results to Ni:Pd bilayers in which the ferromagnetic layer increases the effective Hall coefficient of the non-magnetic layer by a factor of $1 + \chi_m$. Finally, we discuss the implications of these results for earlier studies in rare earth films covered with thin palladium layers. © 2002 Elsevier Science B.V. All rights reserved.

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1. Introduction

In earlier work, two of us used the Hall coefficient to probe hydrogen absorption in gadolinium and dysprosium films [1,2]. In both cases, a rare-earth film was covered with palladium, partly to catalyze atomic hydrogen absorption in the rare earth, partly to stabilize the film mechanically, and partly to protect the rare earth from rapid oxidation in the ambient atmosphere. While the palladium overlayer is much thinner than the rare-earth layer in both cases, its effect on the transport properties of the underlying layer is not trivial, owing to the high resistivities of the rare-earth layers, relative to palladium.

The calculation of the net resistance of two resistors connected in parallel is a common introductory physics exercise. Transferring this exercise to the calculation of the resistance of a bilayer of two parallel layers connected continuously along their length requires replacing macroscopic quantities with their local, microscopic equivalents: resistance with resistivity divided by film thickness, current with current density, and voltage with electric field. The electrical conductivities of the two layers are averaged, with thickness as a weighting factor. But what of the Hall coefficient of such a bilayer?

This question was first answered in 1952 by Nedoluha and Koch [3]. For a film lying in a plane perpendicular to the magnetic field, the composite Hall coefficient of the bilayer equals

$$R_{H3} = \frac{(t_1 + t_2)\left[(R_{H1}\sigma_1^2 t_1 + R_{H2}\sigma_2^2 t_2) + R_{H1}R_{H2}\sigma_1^2\sigma_2^2(R_{H2}t_1 + R_{H1}t_2)B^2\right]}{(\sigma_1 t_1 + \sigma_2 t_2)^2 + \sigma_1^2\sigma_2^2(R_{H2}t_1 + R_{H1}t_2)^2 B^2} \tag{1}$$

where B represents the magnetic field, the subscripts 1, 2, and 3 signify the transport properties of the two constituent layers and the composite bilayer, respectively, R_{Hi}, σ_i, and t_i represent the Hall coefficient, electrical conductivity, and thickness, respectively, of the ith layer. In the limit of small magnetic fields ($\sigma R_H B \ll 1$), this expression reduces to

$$R_{H3} = \frac{(t_1 + t_2)(R_{H1}\sigma_1^2 t_1 + R_{H2}\sigma_2^2 t_2)}{(\sigma_1 t_1 + \sigma_2 t_2)^2} \tag{2}$$

One can interpret Eq. (2) as showing that not only the electric conductivity but also the Hall conductivity ($\sigma_H = \sigma^2 R_H B$ in the small-field limit) is averaged by the measurement process, with thickness acting as a weighting factor:

$$\sigma_3 = \frac{\sigma_1 t_1 + \sigma_2 t_2}{t_1 + t_2} \tag{3}$$

$$\sigma_{H3} = \frac{\sigma_{H1}t_1 + \sigma_{H2}t_2}{t_1 + t_2}$$

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An interesting consequence of this expression is that the composite Hall coefficient of a bilayer is not necessarily intermediate in value to the two Hall coefficients of its constituent layers: the Hall conductivity is averaged, not the Hall coefficient. This is most striking when one of the two layers has a much larger resistivity than the other ($\rho_1 = 1/\sigma_1 \gg \rho_2$). Assuming $R_{H1} \approx R_{H2}$, the maximum Hall coefficient for such a bilayer occurs when the more resistive layer is thicker than the less resistive layer by the ratio $t_1/t_2 = \rho_1/\rho_2$. In this limit, the contribution of the less resistive material to σ_H of the bilayer is larger than its contribution to σ by roughly a factor of ρ_1/ρ_2. Consequently,

$$R_{H3} = \frac{\sigma_{H3}}{B\sigma_3^2} \approx \frac{\rho_1}{4\rho_2} R_{H2} \qquad (4)$$

which can greatly exceed, in magnitude, the Hall coefficient of either constituent layer.

For the particular case of a 10-nm layer of Pd covering a 300-nm-thick Gd film [1], this means that the Hall coefficient of the rare earth could be 1.8 times the Hall coefficient measured for the bilayer. For a 20-nm layer of Pd covering a 300-nm-thick Dy film [2], this means that the Hall coefficient of the Dy layer could be 0.65 times the Hall coefficient measured for the bilayer.

This ignores, however, para- or ferromagnetic effects of the rare-earth layer on the palladium overlayer [1,2]. Gd is a ferromagnet below 293 K [4] and Dy is antiferromagnetic below 179 K [4]. Both are paramagnetic at higher temperatures. One would expect the Hall effect within the Pd film to be enhanced by the ratio of the magnetic field within the Ni film to the applied magnetic field, $B/\mu_0 H = 1 + \chi_m$ (in SI units), where χ_m is the magnetic susceptibility of the rare earth.

The Hall resistivity in a ferromagnet material is given [4] by

$$\rho_H = R_S M + R_0 B \qquad (5)$$

where R_S and R_0 are, respectively, the anomalous and normal Hall coefficients of the material, and M and B are the magnetization and the magnetic field, respectively, which are related to the applied magnetic field, $\mu_0 H$, by

$$B = \mu_0(H + M) \qquad (6)$$

and

$$M = \chi_m H \qquad (7)$$

Thus, the palladium overlayer would experience a larger magnetic field, with the result of enhancing the Hall voltage in that layer by a factor of $(1 + \chi_m)$.

2. Experiment

To test Eqs. (2) and (3), we fabricated unannealed, 100–600-nm-thick bilayers of Zn and Mn, for which
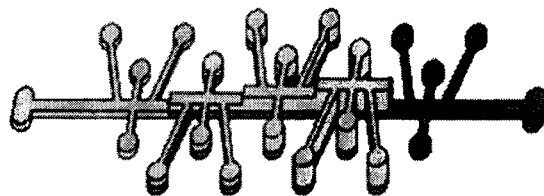

Fig. 1. Specimen geometry. A central horizontal channel is connected to contact pads by thin wires of the same material as the central channel. Four contacts on the top and bottom of each section of the central channel, and the two 'current' electrodes on the left and right ends allow one to measure the resistivity and Hall coefficient of each of five different sections. A computer-controlled shutter allows one to deposit a specimen consisting of five separate bilayers, each with a different combination of layer thicknesses. Typically, the leftmost and rightmost sections are single layers of each material comprising the bilayer. (Vertical scale is greatly exaggerated.)

published resistivities, 5.92 $\mu\Omega$ cm and 136 $\mu\Omega$ cm, respectively [5,6], differ by a factor of 23. Neither material has a sufficiently large magnetic susceptibility to significantly affect the magnetic field experienced by the other. The maximum Hall coefficient for a Zn:Mn bilayer would be approximately six times larger than the Hall coefficient of either metal, according to Eq. (4).

We deposited bilayers on a 75 mm × 25 mm glass substrate using electron beam bombardment in a high vacuum chamber with approximately 10^{-6} torr base pressure. A mask produced the specimen shape shown in Fig. 1. The specimen consisted of a 1-mm-wide central channel connected by thinner side arms to contact pads of the same material, to which electrical connections were made. A computer-driven shutter between the target material and the mask allowed us to simultaneously produce films of five different thicknesses: a single layer each of Mn and Zn at the extremes of the specimen, and three Mn:Zn bilayers between them. We measured layer thicknesses using both a quartz crystal oscillator and an interferometric microscope.

Each bilayer section was connected by thin wires of the same material to six electrical contacts: two 'current' electrodes, shared by all five sections, at the left and right ends of Fig. 1; two 'Hall' electrodes each, connected by short legs perpendicular to the central channel, on opposite sides of the channel; and two 'resistivity' electrodes each, connected to the channel by longer, diagonal legs.

We made Hall measurements in fields of up to 0.6 T, with measurements averaged over both directions of magnetic field and current flow. The Hall angle was less than 1% for all measurements, allowing us to use the low-field limit represented in Eq. (2). We eliminated non-zero offset voltages at zero field electronically. We measured resistivity using the van der Pauw method for the 'current' and 'Hall' electrodes. To test the consisten-
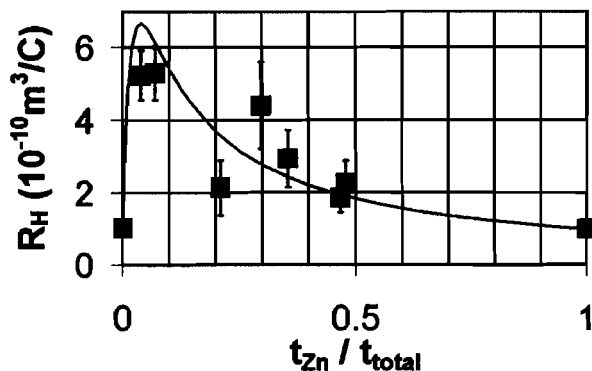
Fig. 2. Hall coefficient of Mn:Zn bilayers vs. fractional thickness of the Zn layer. Data include both Mn ($t_{Zn}/t_{total}=0$) and Zn ($t_{Zn}/t_{total}=1$) single layers. Error bars for the single-layer data are of approximately the same size as the symbols. The line represents the prediction [3] of Eq. (2).

Fig. 4. Hall conductivity of Mn:Zn bilayers vs. fractional thickness of the Zn layer. The line represents the prediction [3] of Eq. (3).

cy of results, we made traditional bar-type resistivity measurements for a few specimens using the 'current' and 'resistivity' electrodes.

To test the effects of a magnetic sublayer on the Hall effect in the covering layer, we evaporated two Ni:Pd bilayers as already described. Each contained five films which could be measured separately for transport properties: one single layer each of Ni and Pd, and three Ni:Pd bilayers of varying thicknesses. The thicknesses of both of the Ni films were constant within each specimen (7 nm and 17 nm) while the thicknesses of the Pd films varied from 100 to 400 nm.

We measured Hall transport for 14 different values of magnetic field up to 0.6 T for nine different films in the two specimens: both single layers of Ni, all six Ni:Pd bilayers, and one single layer of Pd. Finally, we measured Hall transport at 41 different values of magnetic field for four films: each single layer of Ni and one Ni:Pd bilayer from each specimen.
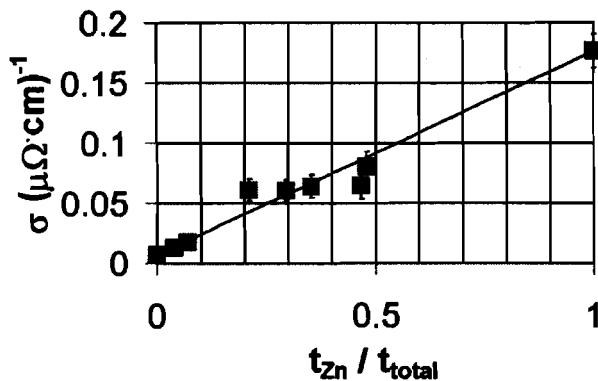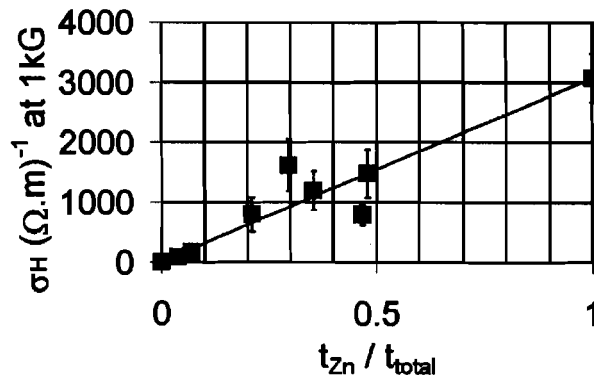
3. Results and discussion

We measured 5.7 ± 0.4 $\mu\Omega$ cm and 140 ± 10 $\mu\Omega$ cm for the resistivity of single layers of Zn and Mn, respectively, and $+1.0 \pm 0.1 \times 10^{-10}$ m^3/C for the Hall coefficient of both. The Hall coefficient for these and other films is displayed in Fig. 2, with the horizontal axis showing the ratio of the thickness of the Zn layer to the total thickness of the bilayer, t_{Zn}/t_{total}. If Eq. (3) is correct, then σ and σ_H should vary linearly with this ratio.

To test this theory, we plotted electric conductivity [Fig. 3], Hall conductivity (Fig. 4), and carrier concentration (Fig. 5) vs. the fractional thickness of the Zn layer. In Figs. 2–5, we also display the prediction of Eqs. (1)–(3), using the values of resistivity and Hall coefficient already measured for pure Zn and Mn. These fits contain no other degrees of freedom. These figures confirm the Nedoluha–Koch model in the Mn:Zn bilayer and demonstrate that the Hall coefficient of the bilayer can exceed its values for the two constituent materials.

While our measured values for resistivity in both Mn and Zn are in excellent agreement with published values,
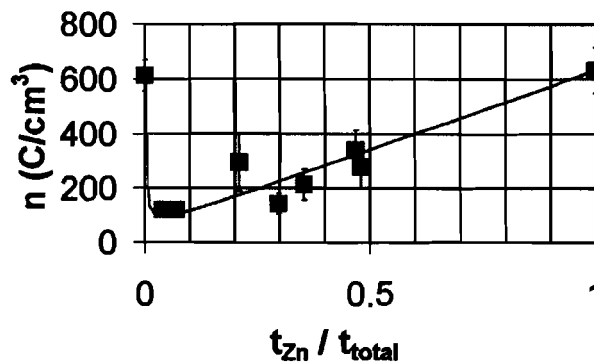

Fig. 3. Conductivity of Mn:Zn bilayers vs. fractional thickness of the Zn layer. The line represents the prediction [3] of Eq. (3).

Fig. 5. Carrier density, $n=1/R_He$, for Mn:Zn bilayers vs. fractional thickness of the Zn layer. The line represents the prediction of Eq. (2).
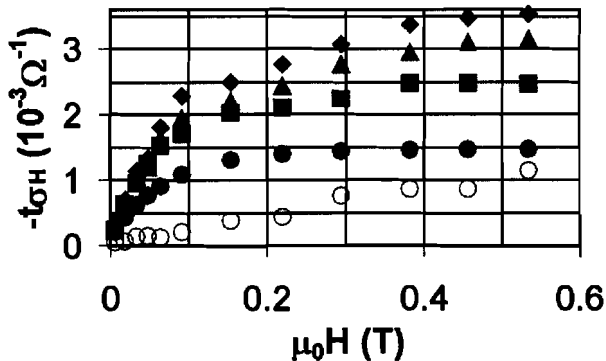
Fig. 6. Thickness times negative of the Hall conductivity for five films in a single specimen of Ni:Pd. From bottom to top: 266 nm layer of Pd; 17 nm layer of Ni; Ni:Pd bilayers with 17 nm Ni thickness and 108 nm, 215 nm, and 266 nm thickness of Pd, respectively.
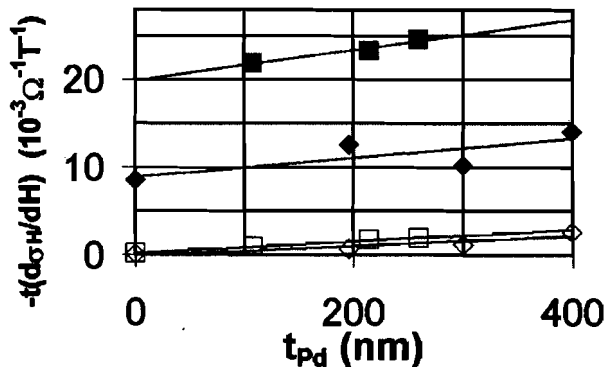

Fig. 7. The high-field (0.4–0.6 T) and low-field (0–0.3 T) slopes of the curves of Fig. 6 (squares) and the other Ni:Pd specimen (diamonds) as a function of the thickness of the Pd layer covering the Ni film. Filled symbols represent low-field data and open symbols represent high-field data. The Nedoluha–Koch model [3] predicts that each set of data should vary linearly with the Pd layer thickness.

and our Hall data for Mn are in fair agreement with Foner's result [7] of $+0.84 \times 10^{-10}$ cm³/C (although in contrast with another published value [5] of 0.93×10^{-10} cm³/C), our Hall measurements for Zn disagree with the published results of $+0.55 \times 10^{-10}$ cm³/C for polycrystalline specimens by Bordin [8] and by Lane et al. [9] We are unable at present to explain this discrepancy.

Our raw data for thickness times Hall conductivity for the covered ferromagnet NiPd system are displayed in Fig. 6, with the slopes of $-t\sigma_H$ vs. applied magnetic field, $\mu_0 H$, for both the low-field (0–0.3 T) and the high-field (0.4–0.6 T) limit are displayed as a function of Pd layer thickness in Fig. 7. The linear fits to each of the four sets of data confirms that $t\sigma_H$ is additive, even though Ni has not yet fully reached its high-field limit at these fields. The slopes of these linear fit lines, which should equal the Hall conductivity of the Pd layer, indicate that the low-field limit of the Hall coefficient of the Pd layers is several times larger (a factor of $1 + \chi_m$, theoretically) than that of a single layer of Pd. ($2.0 \pm 0.4 \times 10^{-10}$ m³/C vs. $0.75 \pm 0.17 \times 10^{-10}$ m³/C) The y-intercepts, which should equal the Ni layer thickness times its Hall coefficient, are consistent with Ni being a ferromagnet.

Next we analyzed the data measured over a range of 41 different magnetic fields to calculate more precisely how the Hall resistivity evolves with increasing magnetic field. We calculated $t\sigma_H$ of the Pd layer of the bilayer by subtracting the values of $t\sigma_H$ in both the Ni and the Ni:Pd films for each data point. We calculated the Hall resistivity for both Ni and Pd layers, which are shown in Fig. 8.

One can calculate the extraordinary Hall coefficient of a ferromagnet by extrapolating the high-field data of Fig. 8 linearly to zero magnetic field. (Since Ni has not yet reached its high-field limit at the magnetic fields

used, this introduces a slight error in this extrapolated value.) For a ferromagnetic material like Ni, the y-intercept equals $\mu_0 M_s R_s$ (SI units), the product of the saturation magnetization, $\mu_0 M_s$, and the extraordinary Hall coefficient. Given that $\mu_0 M_s = 0.609$ T at room temperature [10], we measure $(-52 \pm 1) \times 10^{-10}$ m³/C
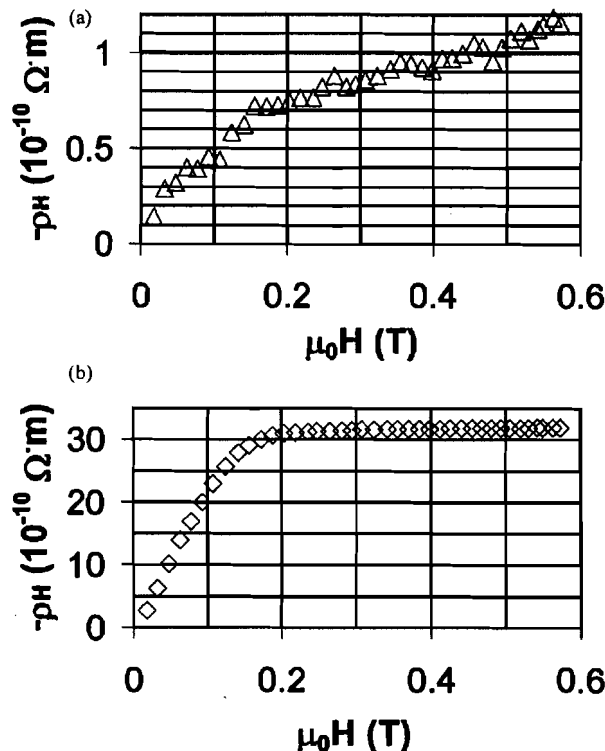

Fig. 8. Hall resistivity vs. applied magnetic field for (a) the Pd layer of the Ni(17 nm):Pd(266) bilayer and (b) the 17 nm Ni single layer of the same specimen.
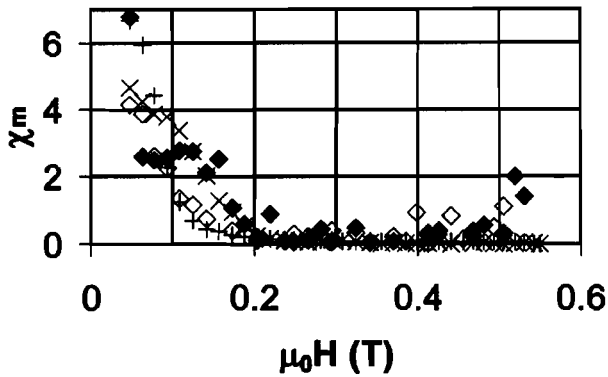
Fig. 9. Magnetic susceptibility as a function of applied magnetic field, as calculated from the Ni single layer ($+$: $t_{Ni} = 7$ nm, $\times$: $t_{Ni} = 17$ nm) and from the Pd layer of a Ni:Pd bilayer (empty diamonds: $t_{Ni} = 7$ nm, solid diamonds: $t_{Ni} = 17$ nm).

for R_s in Ni, in good agreement with Huguenin and Rivier's measurements [11] for polycrystalline films, but somewhat smaller in magnitude than Volkenshtein and Fedorov's measurements [12].

The y-intercept for a Pd film covering Ni (Fig. 8a) is $\mu_0 M_s R_H$, where R_H is the Hall coefficient of Pd. This gives $(-0.64 \pm 0.03) \times 10^{-10}$ m^3/C for R_H, consistent with the -0.68×10^{-10} m^3/C reported in the literature [13]. We measure the high-field slope of ρ_H for this layer to be $(-0.9 \pm 0.1) \times 10^{-10}$ m^3/C and the Hall coefficient of a single layer of Pd to be $(-0.75 \pm 0.17) \times 10^{-10}$ m^3/C.

The derivative $d\rho_H/d\mu_0 H$ tells another story. For Ni, it equals $\chi_m R_s + R_0 \approx \chi_m R_s$ at low fields, while for Pd, it should equal $(1 + \chi_m)R_H$. Thus, we can study how χ_m changes with the field by computing $d\rho_H/d\mu_0 H$, as a function of field for both materials. We have done this, and displayed χ_m in Fig. 9 as a function of $\mu_0 H$. The agreement between Ni and Pd data confirms that the magnetization of the Ni layer does indeed increase the Hall coefficient within the Pd layer by a factor of $1 + \chi_m$, where χ_m is greater than approximately 6 for zero magnetic field.

4. Conclusions

We calculated the product of thickness and Hall conductivity, $t\sigma_H$, as a function of magnetic field for various Mn:Zn bilayers (Fig. 1), in order to test whether the values of this quantity for two layers add when the layers are combined into a bilayer, as predicted by Nedoluha and Koch [3] in Eq. (3).

These results imply that the 10 nm Pd layer of Azofeifa and Clark [1] produces a significant change in the Hall coefficient of a 300 nm Gd film. Assuming 10.55 and 134 $\mu\Omega$ cm for the resistivities of Pd and Gd, and -0.68 and -21×10^{-10} m^3/C, respectively,

for Hall coefficients of Pd [5] and the Gd:Pd bilayer at 23 °C [1], Eq. (2) predicts a Hall coefficient of -38×10^{-10} m^3/C for the Gd itself, 180% of the value measured for the bilayer. Similarly, for the 20-nm Pd layer deposited on 300 nm of Dy in Azofeifa and Clark [2], with a Hall coefficient of -2.0×10^{-10} m^3/C for the bilayer, Eq. (2) implies that the Hall coefficient of the Dy is only -1.3×10^{-10} m^3/C, or 65% of the measured value.

Additionally, since Gd and Dy are ferromagnetic and paramagnetic, respectively, at room temperature, we studied the effect of the internal magnetic field generated by a ferromagnetic Ni layer on the observed Hall effect on a non-ferromagnetic, Pd covering layer. The magnetization of such a material does indeed increase the Hall coefficient within the Pd layer by a factor of $1 + \chi_m$.

For the bulk magnetization $\chi_m = 0.47$ in Gd [1] at room temperature, this magnetization effect implies that the Hall coefficient of the Gd layer is 170%, rather than 180% of that measured for the bilayer. For a bulk magnetization $\chi_m = 0.1$ in Dy [14] at room temperature, this magnetization effect implies that the Hall coefficient of the Gd layer is 50%, rather than 65% of that measured for the bilayer.

In studies of hydrogen absorption in rare earths, Pd overlayers serve to provide mechanical stability, catalyze the absorption of hydrogen, and protect the rare earths from rapid oxidation in the ambient atmosphere. The multi-specimen geometry we describe could allow researchers to eliminate the effects of the Pd overlayer on Hall measurements without having to remove it physically.

Acknowledgments

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