Induced Magnetic Hyperfine Field at Ag Sites near an Fe(100)/Ag(100) Interface

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A magnetic hyperfine field induced at Ag sites near the interface between a Ag(100) crystal and an epitaxially grown bcc Fe(100) film has been measured with monolayer resolution. Utilizing perturbed-angular-correlation spectroscopy with ^{111}In probes a magnetic hyperfine field in the first atomic Ag layer is found to be strongly reduced relative to the Fe bulk value and decreases very quickly as a function of distance from the interface. For both sides of the interface the temperature dependence is well described by a $T^{3/2}$ spin-wave behavior; however, it is strongly enhanced on the nonmagnetic side. [S0031-9007(97)04342-1]

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The magnetic properties of ultrathin films and especially of artificial structures consisting of ferromagnetic layers separated by nonmagnetic spacers have attracted considerable interest in recent years. They have exhibited interesting effects like giant magnetoresistance [1,2] and oscillatory interlayer coupling [3]. Motivated by possible applications in magnetic recording technologies, there has been great effort both experimentally and theoretically to understand the underlying physical principles [4], including detailed Mössbauer studies of the local magnetic hyperfine fields in the ferromagnetic films [5]. In this context, monolayer resolved measurements in the nonmagnetic layers would be important. In this Letter we present first observations of local magnetic hyperfine fields in silver measured by perturbed $\gamma\gamma$ angular correlation spectroscopy.

Very sensitive methods are required for studying monolayer-resolved effects, especially when dealing with buried interfaces. Hyperfine techniques hold great potential here, because the relatively high energy permits the detected radiation to escape the sample without loss of information. Consequently conversion electron Mössbauer spectroscopy (CEMS) at ^{57}Fe probes has been used in the past to investigate Fe films and their interfaces to materials such as Ag, Au, or W [5]. In these experiments about 1 monolayer (ML) of probe atoms is needed to get a sufficiently strong signal. Using perturbed $\gamma\gamma$ angular correlation (PAC) spectroscopy instead overcomes this limitation, because the number of probe atoms is reduced by orders of magnitude to 10^{10}–10^{11} which corresponds to only 10^{-4} ML for common sample dimensions. Isolated radioactive PAC probe atoms can be deposited at any time during sample preparation, thus making it possible to label individual monolayers. These features have permitted the PAC method to be applied very successfully to magnetic surfaces and films in the past [6].

For the experiments reported here, we utilized the isomeric nuclear state ($I = 5/2, t_{1/2} = 84$ ns) of the 171-245-keV $\gamma\gamma$ cascade in ^{111}Cd, which is populated by electron capture of ^{111}In ($t_{1/2} = 2.8$ d). During the lifetime of the isomeric state, the nucleus is subject to interaction between its nuclear magnetic dipole moment [$\mu = -0.7656(25)\mu_N$] and nuclear electric quadrupole moment [$Q = 0.83(13)$ b] with electromagnetic fields produced by the lattice. This leads to an energy splitting of the nuclear level. By coincident observation of the first and second γ quantum, a time dependent angular correlation can be recorded carrying information about the level splitting. The time dependence is characterized by frequencies ω_n, which are the transition frequencies between the nuclear sublevels. For example, the presence of a pure magnetic hyperfine field at ^{111}Cd results in a modulation with the Larmor frequency and its first harmonic, an electric field gradient causes three transition frequencies, and a combination of both acting on the nucleus can cause up to fifteen frequencies. A five-γ-detector setup is used to record simultaneously sixteen coincidence spectra, which are combined to form different counting-rate ratios $R(t)$. Each $R(t)$ can be described as a superposition of cosine and sine modulations with the frequencies ω_n, the corresponding amplitudes depending, for example, on the orientation of the hyperfine fields with respect to the detector setup. Fourier analyses of the $R(t)$ spectra directly exhibit the transition frequencies. However, the hyperfine parameters are obtained by a least-squares fit to all of the time dependent counting-rate ratios. A more detailed description of the experimental method can be found in [7].

Sample preparation and PAC measurements were done using an ultrahigh-vacuum system with a base pressure of 3×10^{-9} Pa. Polished GaAs wafers with (100) surface orientation were first cleaned by Ar$^+$ sputtering at low ion energies, then covered with a 10-ML-thick Fe seed layer to provide the best conditions for epitaxial growth of the following Ag(100) film of several μm thickness [8]. Surface structure was checked using low energy electron diffraction (LEED); possible contaminations were proven to be less than 2% ML in all experiments by means of Auger electron spectroscopy (AES). Best ordered surfaces could be obtained after annealing the Ag film for 60 min at 573 K. Scanning tunneling micrographs of similarly prepared Ag(100) films show atomically flat surfaces with terrace widths of about 30 Å [8].

Three types of samples were produced and measured. The first type of samples was produced with the ^{111}In probes embedded in an Fe film. 25 ML of bcc Fe were epitaxially grown on the Ag(100) surface, then probe atoms were deposited and covered with another 10 ML of Fe. This sample provided calibration values for field strength and temperature dependence inside an Fe film that was thin but nevertheless nearly bulklike. For the second series, isolated carrier-free ^{111}In probe atoms were deposited at room temperature onto a well prepared Ag(100) surface and then covered with 26 ML of Fe. The substrate temperature during deposition was chosen to be either $T_1 = \mathrm{RT}$ or $T_2 = 110$ K. PAC spectra were recorded at various temperatures and for different magnetization directions of the Fe film. For the third type of samples, probe atoms were deposited in the same way but covered with one additional monolayer of Ag before depositing the top Fe film, which in this case had a thickness of 40 ML.

We will first consider the case of probe atoms inside the 35 ML Fe film. After film preparation at $T = 193$ K the sample was magnetized by temporarily applying an external magnetic field of $B \approx 20$ mT. Figures 1(a) and 1(b) show $R(t)$ spectra and corresponding Fourier transforms for two different orientations of the magnetic hyperfine field with respect to the detector plane. The sharp lines in Fig. 1(a) at $\omega_L = 576(1)$ Mrad/s and in Fig. 1(b) at the first harmonic $2\omega_L = 1152(2)$ Mrad/s can be attributed to a magnetic hyperfine field of $|B_{\mathrm{hf}}| = 39.2(2)$ T with $\vec{B}_{\mathrm{hf}}$ parallel to the Fe [100] axes of easy magnetization [9] in agreement with earlier work done on Fe single crystals [10].

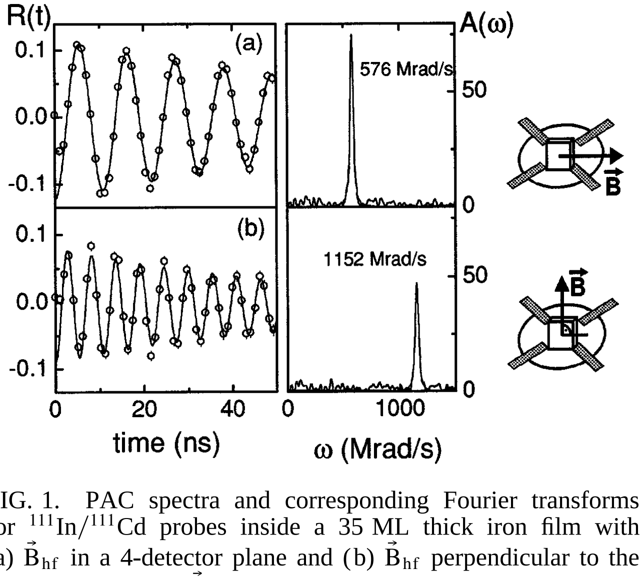

FIG. 1. PAC spectra and corresponding Fourier transforms for ^{111}In/^{111}Cd probes inside a 35 ML thick iron film with (a) $\vec{B}_{\mathrm{hf}}$ in a 4-detector plane and (b) $\vec{B}_{\mathrm{hf}}$ perpendicular to the 4-detector plane. $\vec{B}_{\mathrm{hf}}$ points into the direction of the film magnetization, which has been rotated in an external $\vec{B}$ field (see illustration on the right side; the fifth detector is positioned normal to the 4-detector plane, but not shown for clarity reasons).

We now move on to the more interesting type of samples with ^{111}In deposited at the Fe/Ag interface. Immediately after deposition of the probe atoms onto Ag we observe PAC spectra [Fig. 2(a)] showing very sharp frequencies corresponding to an electric field gradient with quadrupole frequency $\nu_Q = 141.5(1)$ MHz and asymmetry parameter $\eta = 0.00(3)$, in good agreement with earlier measurements on Ag(100) single crystal surfaces [11]. This field gradient is characteristic of the well known substitutional terrace site of ^{111}In in the first monolayer of the Ag(100) surface. After characterizing the sample surface to guarantee good structure and cleanliness, the probes were covered with 26 ML of Fe at a rate of 0.4 ML/min. The substrate temperature during deposition was chosen to be either $T_{\mathrm{dep.}} = \mathrm{RT}$ or $T_{\mathrm{dep.}} = 110$ K. PAC spectra were recorded at temperatures between 110 and 400 K for different magnetization directions of the Fe film. The sample was mounted in such a way that the remnant magnetization direction was parallel to the film plane, but could be turned either parallel to the detector plane or perpendicular to it by temporarily applying an external field without moving the sample (see Fig. 2).

No pronounced oscillation was found after room-temperature deposition of the Fe film, no matter which direction of the magnetic field was chosen. This indicates that interdiffusion hampers the formation of a sharp

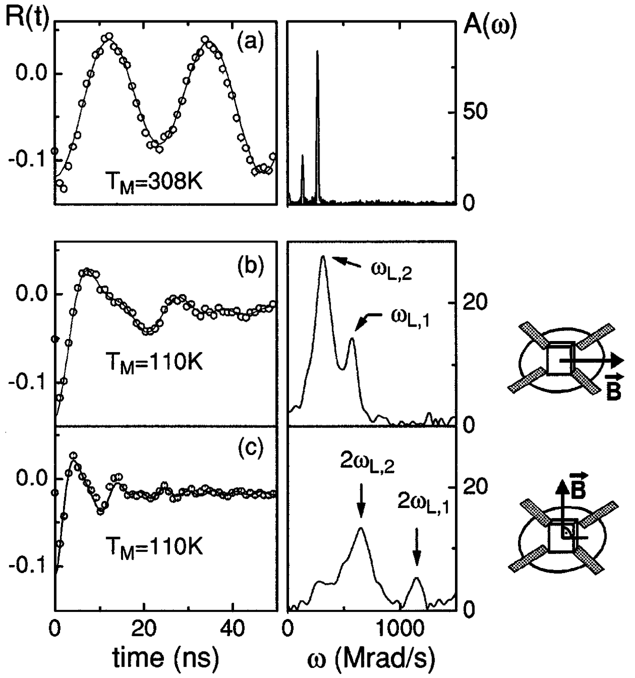

FIG. 2. PAC spectra and corresponding Fourier transforms for ^{111}In probes: (a) at substitutional terrace sites in the topmost atomic layer of a Ag(100) surface (only the first part of numerous oscillations is displayed), (b) after covering the Ag(100) surface with a 15 ML thick iron film with $\vec{B}_{\mathrm{hf}}$ in the 4-detector plane, and (c) same as (b) with $\vec{B}_{\mathrm{hf}}$ perpendicular to the 4-detector plane (see illustration on the right side; the fifth detector is positioned normal to the 4-detector plane, but not shown for clarity reasons).

interface at these temperatures. After deposition at 110 K, on the other hand, clear modulations are observed [Figs. 2(b) and 2(c)] and one can distinguish two major maxima in each Fourier plot. The position of the maxima depends strongly on the field direction and is seen to change by a factor of 2 between the two situations. This proves that the oscillation is mainly a magnetic hyperfine interaction. However, a contribution from electric field gradients is possibly responsible for the observed increased linewidth. Our probe atoms, even when residing in a perfect cubic interface site, are surrounded by two different atomic species, namely Ag and Fe, so the charge symmetry is no longer locally cubic. In addition to that, structural inhomogeneities could give rise to field gradients. Mössbauer experiments with ^{57}Fe probes on the Fe side of the Fe/Ag interface do reveal quadrupolar splittings between 0.005 and 0.064 mm/s [5]. Scaling by the ratio of the nuclear quadrupole moments and Sternheimer antishielding factors for ^{57}Fe and ^{111}Cd, this would translate to quadrupole frequencies up to 70 Mrad/s for our case, which is roughly in the range of our observed linewidths.

The observed frequency peaks can therefore be attributed to two different magnetic probe environments. A least-squares fit applied simultaneously to both $R(t)$ spectra leads to mean values of the magnetic hyperfine field strength of $\vec{B}_{\mathrm{hf},1} = -39.1(3)$ T and $\vec{B}_{\mathrm{hf},2} = -21.6(5)$ T, respectively.

The only reasonable interpretation of these observations is that one site arises from probe atoms in the Fe layer at the interface and the remainder arise from probe atoms in the Ag layer adjacent to the interface. $\vec{B}_{\mathrm{hf},1}$ is almost identical to the value measured before for ^{111}In/^{111}Cd inside the 35 ML iron film (see Fig. 1) and evidently arises from probes in the Fe layer. There is experimental evidence [12] that a certain fraction of atoms exchange places across the interface upon deposition of the Fe film. The adsorption energy of ≈ 3 eV is high enough to allow such processes even at low deposition temperatures. Thus impinging Fe atoms may take the place of some of the ^{111}In probes and cause them to move up one layer into the Fe side. This effect might be enhanced because the metallic radius for In ($r_{\mathrm{In}} = 1.42$ Å) is slightly larger than for Ag ($r_{\mathrm{Ag}} = 1.34$ Å). The first Fe layer ($r_{\mathrm{Fe}} = 1.17$ Å) retains the lattice parameter of Ag and is therefore expanded by 0.8%, which might make it favorable for the In atoms to switch to the Fe side where there is more free space available. Nevertheless, we find only 12(1)% of the probe atoms in the "high field" environment. Further diffusion deeper into the Fe film is kinetically hindered, leaving the probes in the first iron layer where they are exposed to the bulk magnetic field within experimental error. This is consistent with Mössbauer experiments where a 7% enhanced field in the system Ag/Fe(100) was found [5]. This increase is less than the detection accuracy in the PAC experiments described here due to the limiting linewidths.

Most of the probe atoms, 56(2)%, are subject to the reduced field $B_{\mathrm{hf},2}$. These probes have remained in their lattice sites during deposition of the Fe film and are therefore found in the first atomic layer of the Ag film, directly adjacent to the interface with the Fe film. The hyperfine field $B_{\mathrm{hf},2}$ is partly due to the four nearest iron neighbors, but the high value of 21.6(3) T is a strong argument that the silver conduction electrons are polarized, leading in turn to a polarization of the s electrons of the ^{111}Cd probes. A summary of the observed magnetic hyperfine fields across the interface is given in Fig. 3.

Additional confirmation for the above interpretation can be obtained from the temperature dependence of the measured magnetic hyperfine fields, which are all shown in Fig. 4. The accessible temperature range was limited to ≥ 110 K by the capability of the cooling system and to a maximum of 400 K to avoid interdiffusion effects that destroy the layer system. Ahmad *et al.* [13] used classical spin-wave theory to show that a pseudo $T^{3/2}$ behavior of the form $B_{\mathrm{hf}} = B_{\mathrm{hf}}(0)\,(1 - bT^{3/2})$ is applicable to thin films as well as to surfaces and interfaces. In order to demonstrate this behavior, the data in Fig. 4 are plotted versus a $T^{3/2}$ temperature scale and are normalized to $B_{\mathrm{hf}}(0)$ obtained by a least-squares fit (dotted lines). The yielded parameters are summarized in Table I. It is especially interesting to compare the spin-wave parameters b for the PAC and Mössbauer experiments, which are also included in Table I. There is very good agreement within experimental errors between the two types of measurements with probes inside the film or in the first magnetic layer, respectively. Rado [15] and Ahmad *et al.* [13] show that the parameter b_S at surfaces and interfaces should be about twice the bulk parameter b_V. This is consistent with our findings giving further support to our identification of $B_{\mathrm{hf},2}$ as probe sites in the first atomic layer of the Fe film.

The further increased spin-wave parameter for ^{111}In at the nonmagnetic side of the interface of course has no

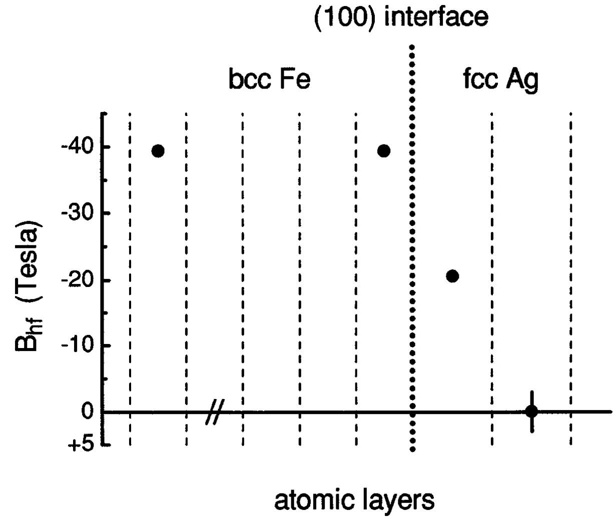

FIG. 3. Magnetic hyperfine field $\vec{B}_{\mathrm{hf}}$ at ^{111}In/^{111}Cd probe nuclei as a function of position relative to the Fe/Ag interface.

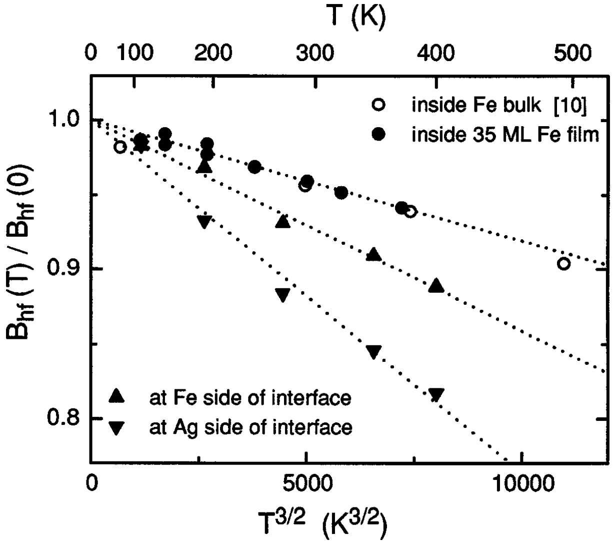

FIG. 4. Normalized temperature dependence of magnetic hyperfine fields measured at ^{111}In/^{111}Cd in the Fe/Ag(100) system. The dotted lines are least-squares fits based on $B_{\mathrm{hf}}(T) = B_{\mathrm{hf}}(0)(1 - bT^{3/2})$.

counterpart in ^{57}Fe experiments. Here the advantage of using very dilute probe atoms is most obvious. The parameter is 67% larger in the Ag layer than in the Fe layer, which is far beyond experimental error.

A nonvanishing magnetic field at the second and third Ag monolayers has been predicted, for example, by Dederichs *et al.* for the system fcc Fe/Cu(100) [16]. We attempted to determine such a field in the second monolayer experimentally. ^{111}In probe atoms were deposited as described above in the topmost monolayer of a Ag film and then covered with one additional monolayer of Ag at $T = 173$ K. After that a relatively thick (40 ML) Fe film was deposited on top at the same substrate temperature. The low temperature is necessary to prevent probe atoms from floating on the Ag film during Ag deposition. After Fe deposition we found a slight change in the PAC signal. However, there was no significant dependence on the direction of a temporarily applied external field. We can only estimate an upper limit of ~3 T for the magnetic hyperfine field strength, because $R(t)$ is damped quite strongly. This does not necessarily contradict calculations, because the field strength is expected to decrease as $1/n^2$ for a single interface with n being the distance to the interface in atomic layers.

TABLE I. Parameters for the temperature dependence of magnetic hyperfine fields at ^{111}Cd and ^{57}Fe in the Fe/Ag system expressed in the form $B_{\mathrm{hf}}(T) = B_{\mathrm{hf}}(0)(1 - bT^{3/2})$.

Probe position	Probe nucleus	$B_{\mathrm{hf}}(0)$ (T)	b (10^{-5} $\mathrm{K}^{-3/2}$)
Inside Fe film	^{111}Cd	−39.9(1)	0.81(6)
(~30 ML)	^{57}Fe [14]	−33.95	0.62
First Fe layer	^{111}Cd	−40.1(1)	1.4(1)
at Fe/Ag	^{57}Fe [14]	−34.92	1.33
First Ag layer			
at Fe/Ag	^{111}Cd	−20.9(2)	2.4(2)

In this Letter we have shown conclusively that a ferromagnetic bcc-Fe film epitaxially grown onto Ag(100) induces a magnetic polarization in the first monolayer of silver. The observed magnetic hyperfine field is relatively strong and vanishes rapidly at deeper layers. The behavior may be different for Fe/Ag/Fe sandwich structures or multilayers where higher magnetic fields are expected in the nonmagnetic spacer due to quantum well effects. The induced magnetic hyperfine field in the first Ag monolayer, in addition, shows the striking feature of a rather strong temperature dependence. In order to understand the observed magnetic effects further theoretical calculations would be extremely important.

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[1] M. N. Baibich, J. M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Eitenne, G. Creuzet, A. Friederich, and J. Chazelas, Phys. Rev. Lett. **61**, 2472 (1988).
[2] G. Binasch, P. Grünberg, F. Saurenbach, and W. Zinn, Phys. Rev. B **39**, 4828 (1989).
[3] S. S. P. Parkin, N. More, and K. P. Roche, Phys. Rev. Lett. **64**, 2304 (1990).
[4] *Ultrathin Magnetic Structures II,* edited by B. Heinrich and J. A. C. Bland (Springer-Verlag, Berlin-Heidelberg, 1994).
[5] G. Liu and U. Gradmann, J. Magn. Magn. Mater. **118**, 99 (1993).
[6] G. Krausch, R. Fink, K. Jacobs, U. Kohl, J. Lohmüller, B. Luckscheiter, R. Platzer, B.-U. Runge, W. Wöhrmann, and G. Schatz, Hyperfine Interact. **78**, 261 (1993).
[7] G. Schatz and A. Weidinger, *Nuclear Condensed Matter Physics* (Wiley, Chichester, 1996).
[8] D. E. Bürgler, C. M. Schmidt, J. A. Wolf, T. M. Schaub, and H.-J. Güntherodt, Surf. Sci. **366**, 295 (1996), and references therein.
[9] J. J. Krebs, Appl. Phys. A **49**, 513 (1989).
[10] See, for example, B. Lindgren and Y. K. Vijay, Hyperfine Interact. **9**, 379 (1981).
[11] R. Wesche, R. Fink, T. Klas, G. Krausch, R. Platzer, J. Voigt, and G. Schatz, J. Phys. Condens. Matter **1**, 7407 (1989).
[12] W. F. Egelhoff, Jr., Mater. Res. Soc. Symp. Proc. **229**, 27 (1992).
[13] B. S. Ahmad, J. Mathon, and M. S. Phan, J. Phys. Colloq. (France) **49**, 1639 (1988).
[14] J. Korecki and U. Gradmann, Europhys. Lett. **2**, 651 (1986).
[15] G. T. Rado, Bull. Am. Phys. Soc. **2**, 127 (1957); G. T. Rado and J. C. Walker, J. Appl. Phys. **53**, 8055 (1982).
[16] P. H. Dederichs, P. Lang, K. Willenborg, R. Zeller, N. Papanikolaou, and N. Stefanou, Hyperfine Interact. **78**, 341 (1993).