Origin of Biquadratic Coupling in Fe/Cr(100) Superlattices

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We investigate the magnetic properties of a (100) oriented $[\mathrm{Fe}(1.7\ \mathrm{nm})/\mathrm{Cr}(8.4\ \mathrm{nm})]_{10}$ superlattice by means of perturbed angular correlation spectroscopy. The magnetic ordering in the Cr layers is obtained by measuring the magnetic hyperfine interaction at implanted ^{111}Cd nuclear probes. We identify dynamic antiferromagnetic spin fluctuations in the Cr layers and show that it gives rise to the biquadratic interlayer coupling.

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Dynamical effects play a ubiquitous role in diverse solid-state phenomena. The experimental challenge is to identify suitable probes that span the time scales of interest. In the present work, we introduce perturbed angular correlation (PAC) spectroscopy as a novel technique to study dynamical phenomena in nanostructures. We complement previous neutron diffraction studies of Fe/Cr superlattices and address an open basic question about magnetic coupling between thin metallic films — the microscopic origin of the non-Heisenberg, biquadratic coupling. We show that biquadratic interlayer coupling, i.e., the coupling that orients adjacent Fe layers 90° apart, is mediated by superparamagnetic Cr moments that fluctuate in the GHz regime.

The discovery of interlayer coupling between ferromagnetic layers across nonferromagnetic spacer layers [1] stimulated much research. It was first recognized that the coupling in Fe/Cr multilayers aligns adjacent Fe layers parallel or antiparallel, depending on the Cr thickness, with a Cr thickness periodicity of 1.8 nm [2]. Then, an additional oscillation period of two monolayers was found [3]. Finally, a biquadratic contribution to the interlayer coupling was discovered that favors perpendicular alignment of adjacent Fe layers [4]. The theoretical explanation for the long- and the short-period oscillatory coupling depends on the electronic properties of the spacer [5]. However, such theories lead only to parallel or antiparallel alignment of the ferromagnetic layers and do not describe the biquadratic coupling. While various mechanisms have been proposed to explain the existence of biquadratic coupling [6], experimentally, its origin remains to be clarified.

It is of fundamental importance to understand the interplay between the intrinsic Cr spin density wave (SDW) magnetism [7] and the interlayer coupling in the model system Fe/Cr. The magnetic ordering of Cr spacer layers is expected to depend on the structure of the Cr [8] and of the Fe/Cr interface [9]. One should therefore make a clear distinction between the results obtained on nearly ideal Fe whiskers and those on GaAs, Al_2O_3, or MgO substrates. The latter is studied herein. In 1995, Fullerton *et al.* showed that antiferromagnetic (AF) ordering of Cr dramatically alters the interlayer coupling between the Fe layers of Fe/Cr superlattices [10]. The biquadratic interlayer coupling observed in the thick Cr regime ($t_{\mathrm{Cr}} \geq 5.0$ nm) is suppressed below the Néel temperature (T_N) of the Cr spacer. PAC measurements identified SDW ordering in the Cr layers, and confirmed the SDW instability below a critical thickness [11]. It is now clear that, in superlattice systems, the long-range SDW ordering in the Cr spacer destroys the interlayer coupling. Later, neutron diffraction measurements identified a gradual transition from incommensurate to commensurate AF order in the temperature range between 175 and 310 K and to paramagnetic Cr at $T_N = 500$ K [12]. However, the nature of the microscopic magnetic ordering in the transitional region and its relation to the interlayer coupling still remains unclear [13]. In this Letter, we will identify dynamical effects in Cr. This information is essential to understand the phenomenon of biquadratic coupling.

The $[\mathrm{Fe}(1.7\ \mathrm{nm})/\mathrm{Cr}(8.4\ \mathrm{nm})]_{10}$ superlattice was grown at 310 °C on a MgO(100) substrate by means of molecular beam epitaxy (MBE) with a base pressure of 5×10^{-11} Torr. X-ray diffraction and Rutherford backscattering experiments indicate high quality epitaxial growth. The temperature dependent resistivity shows an anomaly that is smeared out between 175 and 300 K. We obtain a value of (200 ± 5) K for T_N from the minimum in the derivative of the resistivity. Our value agrees with that for sputtered superlattices [10] and other MBE-grown superlattices [12]. Magnetization and magnetoresistence data were recorded with the magnetic field **H** applied in plane along the Fe [001] easy axis. We obtained similar results to those reported by Fullerton *et al.* on an $[\mathrm{Fe}(1.4\ \mathrm{nm})/\mathrm{Cr}(7.0\ \mathrm{nm})]_{13}$ superlattice [10]. We recorded a maximum magnetoresistance $\Delta\rho/\rho = 0.62\%$ at 200 K. The temperature dependence of the saturation field, shown in Fig. 1, indicates that the interlayer coupling is suppressed below 200 K. A study of similarly grown sandwiches [14] and the results of Fullerton *et al.* lead us to conclude that our superlattice shows biquadratic coupling above 200 K and vanishing coupling below that temperature. In the following, PAC spectroscopy is used to probe the local magnetic ordering in the Cr layers.

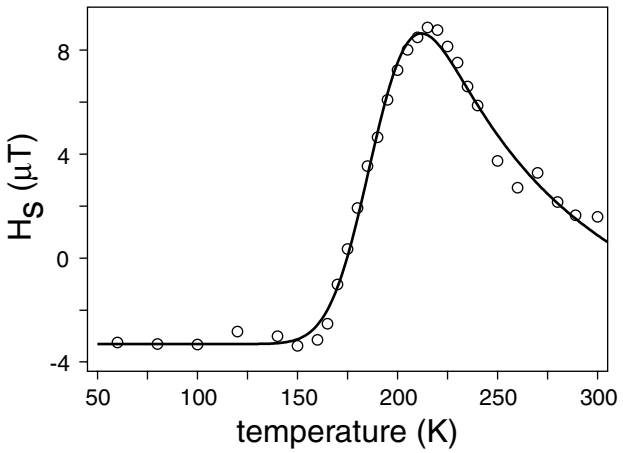

FIG. 1. Saturation field H_S vs temperature. H_S is defined as the field where the remanent magnetization is 90% of the saturation magnetization. The line is a fit using the loose-spin model [23] with potential $|U/k_B| = 8$ K, and using a Gaussian distribution (mean = 193 K, width = 17 K) of blocking temperatures below which the biquadratic coupling is suppressed.

In a PAC experiment, one measures the angular correlation between two photons successively emitted in the γ-γ cascade of ^{111}Cd (the daughter nucleus of ^{111}In) [15]. During the lifetime of the intermediate nuclear level, the hyperfine fields exert a torque on the moments of the probe. This torque can cause a reorientation of the nuclear spin (similar to the Larmor precession) which affects the emission probability of the second γ ray. If one looks in a particular direction, one essentially observes an exponential decay perturbed by the hyperfine interaction. Coincidence spectra W_{90} and W_{180} are measured with detectors at 90° and 180°, respectively. From these, the time dependent anisotropy function $R(t) = 2(W_{180} - W_{90})/(W_{180} + 2W_{90})$ is constructed. The $R(t)$ spectrum (known also as the PAC time spectrum) contains the information on the hyperfine interactions (frequencies $\nu = \omega/2\pi$ and absolute fractions). For the ^{111}Cd probes the frequency is proportional to the hyperfine field by the factor 2.33 MHz/T.

The anisotropy function can be theoretically simulated by calculating the time evolution of the different quantum states in the intermediate nuclear level under the influence of the hyperfine interaction Hamiltonian [15]. In the case of ferromagnetic or commensurate AF (AF_0) ordering, the general expression reduces analytically to $R(t) = a_0 + a_1 \cos(\omega t) + a_2 \cos(2\omega t)$. The values a_0, a_1, and a_2 depend on the orientation of the hyperfine field with respect to the detectors, with $a_0 + a_1 + a_2 = A_{22}$, a nuclear constant. In the case of an incommensurate SDW ordering (AF_1 or AF_2) one has to account for the Overhauser distribution with cutoff frequency ω_0 which leads to $R(t) = a_0 + a_1 J_0(\omega_0 t) + a_2 J_0(2\omega_0 t)$, with J_0 being the Bessel function of zeroth order, and the amplitudes having the same angular dependence as in the commensurate case.

For the PAC measurements, 2×10^{13} atoms/cm^2 of radioactive ^{111}In were implanted at 80 keV with the sample at room temperature and slightly tilted to avoid channeling. X-ray diffraction spectra and the magnetization were measured before and after implantation with no measurable differences. Experimental PAC data were recorded in the configuration as shown in Fig. 2(a). The three spectra are consistently analyzed with 17% of the substitutional nuclear probes located in the Fe layer and 83% in the Cr. Probe atoms at the interfaces and those in ill-defined environments are responsible for the rapid (<2 ns) reduction of the anisotropy. The rapid oscillations in the spectra reflect the Fe signal. The measured hyperfine interaction

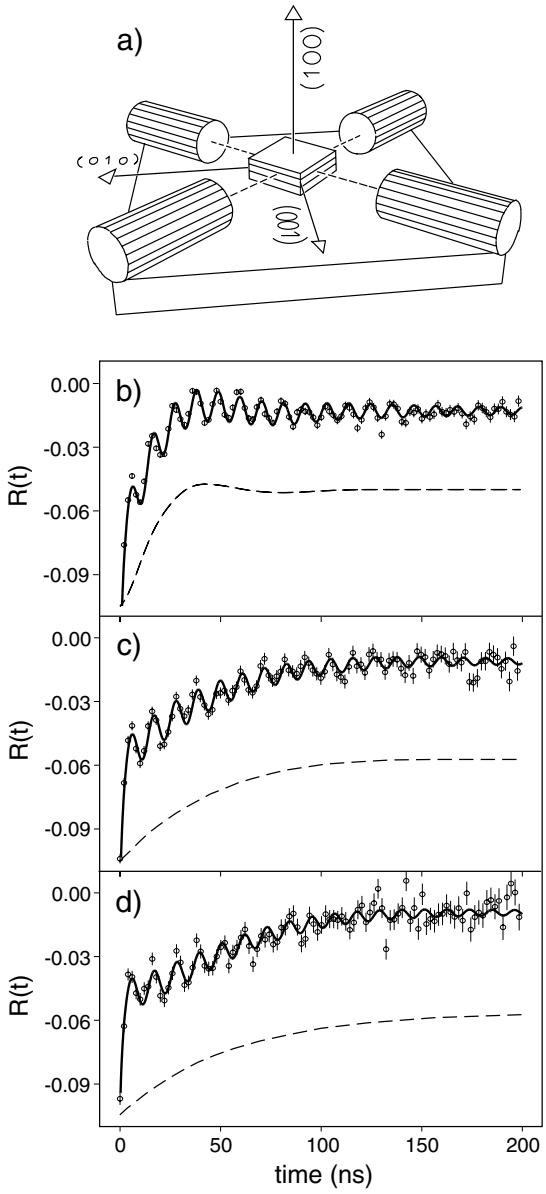

FIG. 2. Configuration of the sample with respect to the detectors (a). PAC time spectra obtained at 77 (b), 225 (c), and 300 K (d). The solid lines are fits described in the text. The dashed lines represent the chromium contribution.

frequencies $\nu = 92.5(2)$, 90.9(2), and 89.3(2) MHz correspond to the bulk values at 77, 225, and 300 K, respectively. The well-known behavior of in-plane magnetization for Fe is confirmed since only the single frequency of the Fe-hyperfine field is observed.

For the PAC time spectrum recorded at 77 K, Fig. 2(b), the Cr contribution consists of a damped Bessel function with frequency $\nu = 12.2(2)$ MHz, and therefore indicates SDW ordering in the Cr layers. As explained in [16], the small hyperfine field value is indicative of a transverse SDW (AF_1). Because only the first harmonic of the Bessel function is observed, the Cr magnetization is oriented in plane. The observation of a transverse SDW is at variance with our previous studies [11]. However, investigation of samples grown at different temperatures shows that the enhanced value of T_N and the occurrence of a longitudinal SDW (AF_2) observed in earlier work was related to the reduced growth temperature (between 0 and 50 °C). These findings not only explain the discrepancy between results of various groups [12,17], but also indicate that it is possible to *control* the type of SDW by varying the growth conditions.

The PAC time spectra recorded at 225 K and at 300 K are shown in Figs. 2(c) and 2(d). The Cr signal obtained here consists of an exponential-like decay. This signal is obviously of magnetic origin, but cannot be analyzed in terms of a static commensurate, incommensurate, or helical SDW [13], as these structures would lead to a pronounced precession pattern in the PAC time spectrum [16]. The interface contribution consists of a steeply decaying spin polarization of Cr with a penetration depth of about 0.1 nm [18], but it cannot explain the observed spectra. In the regime where biquadratic interlayer coupling occurs, the PAC results are incompatible with static magnetic ordering in the Cr.

It is known that an exponential attenuation of the anisotropy may result from dynamical effects. We evaluated the anisotropy function following the procedure developed by Winkler and Gerdau [19] on the basis of Blume's stochastic model [20]. Using this stochastic model, the anisotropy function is completely determined by the various stochastic states and by their characteristic lifetime τ. We consider in Fig. 3 the situation in which the hyperfine field can jump between two opposite directions that are each 45° from a detector. This means that the magnetization jumps between positive and negative in-plane [001] axes, as shown in Fig. 2. Since we are concerned with the Cr magnetization, we account for the Overhauser distribution by a weighted sum taken over 1000 points in the frequency domain. For very long characteristic times ($\tau \gg 1/\omega_0$), the anisotropy function reduces to the statistical average of the anisotropy functions for the individual, static hyperfine fields. Therefore, we obtain in the slow-fluctuation limit the Bessel function of zeroth order. For this orientation of the magnetic fields only the single frequency is present ($a_0 = a_2 = 0$).

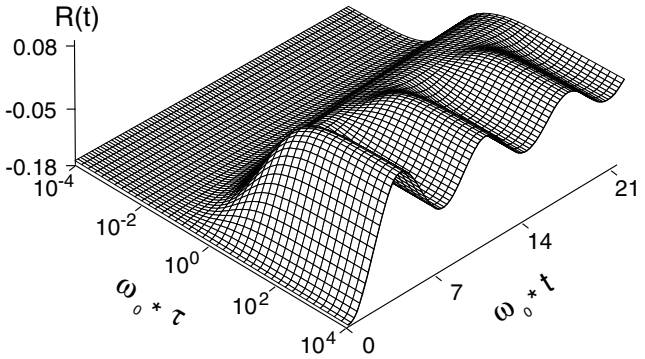

FIG. 3. Simulation of PAC spectra using the stochastic theory as described in the text.

When the time scale of the fluctuation matches that of the characteristic frequency of the hyperfine interaction, the spectrum is that of an overdamped oscillator. For the limiting case of very fast fluctuations we expect the nucleus to see the time-averaged magnetic hyperfine field, a phenomenon known as motional narrowing in the context of line-shape studies. In our case the time-averaged field is zero, therefore the fast-fluctuation limit results in a paramagneticlike spectrum.

The Cr signal in the PAC spectra of Fig. 2 is well reproduced using the stochastic model. The three spectra are analyzed consistently with only the hyperfine field value for the Fe site and the characteristic lifetimes for Cr as fitting parameters. We obtain for the spectra at 225 and 300 K the values of $\tau = 2.2$ ns and $\tau = 0.6$ ns, respectively. Relating this information to the coupling behavior derived from Fig. 1, it becomes evident that fluctuating Cr magnetic moments coexist with the biquadratic interlayer coupling in the Fe/Cr superlattice.

The PAC data disclose superparamagnetic Cr with moments fluctuating in the GHz regime. It is interesting to note that Fawcett *et al.* recently considered the phenomenon of superparamagnetic relaxation in Cr and recognized the relevance to Fe/Cr superlattices [21]. The fluctuating magnetic behavior is not restricted to small clusters around diluted nucleation centers (i.e., Fe impurities), as this would lead to a large paramagnetic signal in the PAC spectra superimposed on the signal arising from magnetic environments. Also, the PAC spectra rule out the possibility of *static* commensurate, incommensurate, or helical AF ordering along with the biquadratic interlayer coupling in this thickness and temperature regime. On the other hand, a local probe technique provides only limited information on the lateral coherence length when dynamical effects are present. Theoretical calculations predict commensurate AF order [13] and the formation of domains in the antiferromagnet [22] at elevated temperatures. We verified that a model of superparamagnetic AF_0 Cr, although not fitting the data at 77 K, is able to fit the spectra at 225 and 300 K. The values for the characteristic times depend slightly on the chosen stochastical model. The

analysis in terms of superparamagnetic Cr with polarization fluctuations in the plane and between two opposite directions is consistent with the in-plane AF ordering obtained via neutron diffraction [12]. Whereas PAC measurements are most sensitive to spin correlation times τ between 10^{-10} and 10^{-8} s, neutron diffraction is most sensitive to fluctuations in the time range of 10^{-12} to 10^{-14} s. Therefore, AF ordering with fluctuations in the time range of 1 ns will appear as static in neutron diffraction data. Moreover, neutron diffraction and nuclear techniques sample different regions of $\mathbf{q}$ space, and these regions exhibit different dynamical behavior.

The observed microscopic magnetic ordering in the Cr layers is reminiscent of the loose-spin mechanism proposed by Slonczewski to explain biquadratic interlayer coupling [23]. In the loose-spin model, localized spins in the spacer layer contribute an exchange coupling between the ferromagnetic films. The contribution to the free energy that favors biquadratic coupling is derived from conventional statistics. In many cases, the theory produces the correct experimental temperature dependence for the biquadratic interlayer coupling strength [24,25]. The model reproduces also the strong temperature dependence of the coupling strength above the blocking temperature of the Cr (Fig. 1). Our PAC results provide *direct* evidence of fluctuating localized spins in the Cr layer when the Fe layers are biquadratically coupled. In the model chosen to fit the PAC data, the Cr spins fluctuate between two opposite directions with a time-averaged moment of zero. In this respect, the analysis is at variance with the loose-spin model. It is expected that more sophisticated stochastic models may fit the PAC spectra as well. Yet, in spite of the simplifications, the present model successfully describes SDW ordering in Cr below, and dynamical behavior in Cr above the blocking temperature. It may also be interesting to refine the loose-spin model by incorporating fluctuations of the loose spins between two opposite states. Probably, bilinear coupling cancels out naturally when the first moment of the magnetization of the loose spins vanishes, thereby making the assumption of spacer layer thickness fluctuations unnecessary to explain the dominance of biquadratic coupling in some systems.

In conclusion, we determined the magnetic properties of an MBE-grown MgO/[Fe(1.7 nm)/Cr(8.4 nm)]$_{10}$ superlattice. Macroscopic measurements reveal a critical temperature for the Cr of 200 K, above which the Fe layers are biquadratically coupled. Perturbed angular correlation spectroscopy is used to observe the hyperfine interactions on implanted radioactive nuclei. At low temperatures, a SDW ordering in Cr is measured. At elevated temperatures, above the blocking temperature, superparamagnetic relaxation in Cr is observed. We thus demonstrate that biquadratic interlayer coupling in Fe/Cr goes hand in hand with fluctuating magnetic moments in the Cr spacer layers. It becomes now possible to study the energetics of the fluctuations through the systematic measurement of the characteristic times as a function of temperature and applied field. Alternatively, we expect that the spin dynamics of the spacer can be observed via inelastic neutron diffraction in the present system, or with Mössbauer spectroscopy in the systems Fe/Ag^{57}FeAg/Fe or Fe/^{57}FeSi/Fe. We also suggest that there is a need to analyze the line broadening in the Mössbauer results for Fe/Cr^{119}SnCr/Fe multilayers [26] in terms of superparamagnetic behavior in the Cr layers.

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