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Field-cooling effect in biquadratically interlayer-coupled Fe/Cr/Fe trilayers

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We report on the modified magnetic properties of an epitaxial Fe/Cr/Fe trilayer when it is cooled in an applied magnetic field. Magnetization measurements are carried out to investigate the dependence of the field-cooling effect on the magnetic field strength and on the temperature. Perturbed angular correlation experiments are performed to study the microscopic origin of the field-cooling effect.

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The observation of antiferromagnetic interlayer coupling[1] and of the giant magnetoresistance effect[2] motivated much research on Fe/Cr multilayers. The antiferromagnetic interlayer coupling strength in Fe/Cr multilayers varies as a function of the Cr spacer layer thickness with a two-monolayer period oscillation[3] that is superimposed on a decaying oscillation with a period of 1.8 nm.[4] When the antiferromagnetic interlayer coupling is weak, e.g., for a Cr thickness larger than 5 nm, the biquadratic coupling may lead to an orthogonal alignment of the magnetization in neighboring Fe layers. Much effort was done to investigate the origin of the biquadratic interlayer coupling.[5]

It was found that the biquadratic interlayer coupling in Fe/Cr multilayers strongly depends on the magnetic properties of the spacer layer.[6] Below the Néel temperature of Cr,[7,8] the biquadratic interlayer coupling in Fe/Cr multilayers is suppressed. Above the Néel temperature, dynamical magnetic moments mediate the biquadratic interlayer coupling.[9] In spite of the large efforts to study the biquadratic interlayer coupling in Fe/Cr multilayers, the phenomenon is still not fully characterized.[10] In this work, we report on the observation that the biquadratic interlayer coupling in an Fe/Cr (5.5 nm)/Fe trilayer apparently may be suppressed at low temperatures by cooling the sample in an applied magnetic field.

The MgO(001)/Fe (7.0 nm)/Cr (5.5 nm)/Fe (2.5 nm) sample is grown via molecular beam epitaxy (base pressure of 1.3×10^{-8} Pa) at 450 K. A Cr capping layer 3.5 nm thick, deposited at room temperature, is used to protect the multilayer against oxidation. The epitaxial growth and the layer thicknesses are verified via x-ray diffraction and Rutherford backscattering experiments. The magnetization measurements are performed with a vibrating sample magnetometer and with the magnetic field applied along the in-plane easy axis of the Fe layers.

In Fig. 1(a), we show the magnetization curve recorded on the Fe/Cr (5.5 nm)/Fe trilayer at 10 K. The reduction of the normalized remanent magnetization (M_R/M_S=0.73) is indicative of the biquadratic interlayer coupling. The sample shows biquadratic interlayer coupling from 4 K up to room temperature. The saturation field, defined as the field at which the magnetization reaches 90% of the saturation magnetization, decreases monotonically with increasing temperature from $\mu_0 H_s$=20 mT at 10 K to 12 mT at 290 K.

The magnetization curve obtained after cooling the Fe/Cr (5.5 nm)/Fe trilayer to 10 K in an applied magnetic field of $\mu_0 H$=60 mT is shown in Fig. 1(b). In the first quarter of the hysteresis curve, the normalized remanent magnetization equals unity, indicating that the layers do not order orthogonally. The suppression of the orthogonal ordering, observed in the first quarter of the magnetization loop, is not reproduced in the other quarters of the hysteresis loop. Also, it is not reproduced when a second hysteresis loop is measured without changing the temperature. Apart from the behavior in the first quarter of the hysteresis loop, we could not detect any other differences in the hysteresis loop as a result of field cooling—in particular, the loop is not shifted horizontally and the coercivity is not increased. The absence of the orthogonal ordering in the first quarter of the hysteresis curve after cooling the sample in an external magnetic field will be referred to as the field-cooling effect. Observing the remanent magnetization, one can readily discriminate whether the field-cooling effect is present or not.

In Fig. 2(a), we plot the remanent magnetization measured at T_{meas}=10 K after field cooling the sample from different temperatures T_{FC}. If the sample is field cooled in a field of 100 mT from T_{FC}=200 K to T_{meas}=10 K, then the field-cooling effect occurs and the remanent magnetization equals unity. If, however, the sample is field cooled in a field of 100 mT from T_{FC}=40 K to T_{meas}=10 K, then no field-cooling effect is observed and the remanent magnetization is 0.73. Figure 2(a) thus indicates that the field-cooling procedure must be initiated at a high enough temperature. We will indicate the temperature from above which the sample must

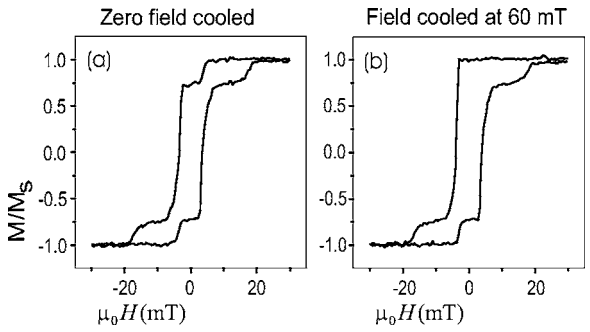

FIG. 1. Hysteresis loops measured at 10 K, (a) zero field cooled and (b) field cooled from 200 K to 10 K at $\mu_0 H$=60 mT.

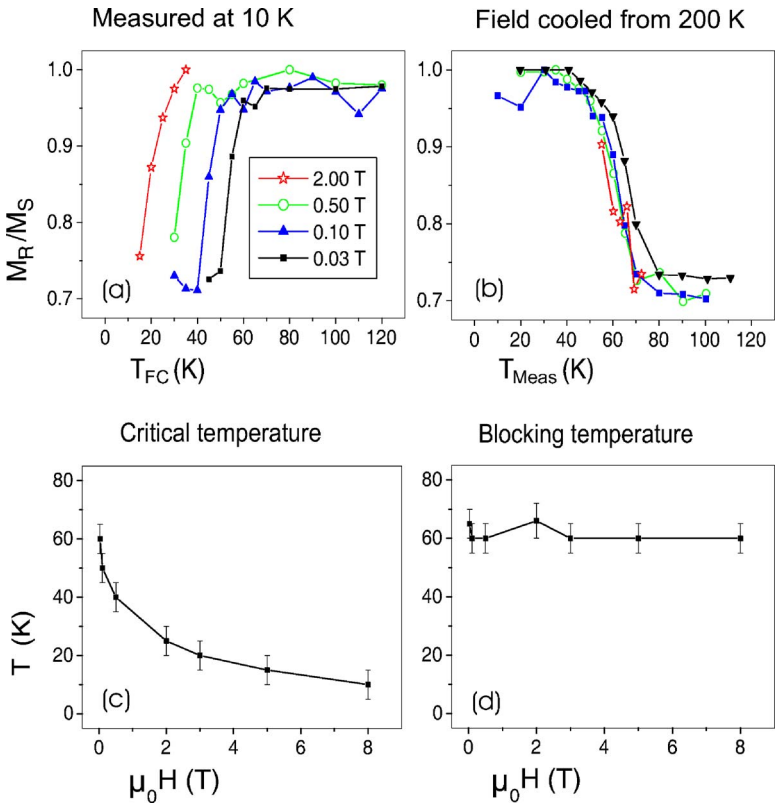

FIG. 2. (Color online) Normalized remanent magnetization M_R/M_S as a function of (a) the temperature from which the field cooling is started and (b) the temperature to which the sample is field cooled starting from 200 K. The sample is field cooled at different applied fields as indicated. (c) The critical temperature as a function of applied magnetic field. (d) The blocking temperature as a function of applied magnetic field.

be field cooled in order to observe the field-cooling effect as the *critical temperature*.

Figure 2(a) shows that the critical temperature depends on the strength of the magnetic field during cooling. The dependence of the critical temperature on the cooling field is plotted in Fig. 2(c). The results show that the critical temperature may even attain the measuring temperature of 10 K when the external magnetic field is 8 T. Indeed, even if the sample was cooled in zero field to 10 K, we could achieve suppression of the orthogonal ordering in the first quarter of the hysteresis curve through the application of a strong magnetic field of 8 T at 10 K. Accordingly, the shape of the hysteresis loop at the first and third quarters may vary, depending on the maximum field that is used for the magnetization measurements.

In Fig. 2(b), we investigate at which temperatures the field-cooling effect may be observed. The sample is repeatedly field cooled from $T_{FC}=200$ K to the temperature at which the hysteresis loop is measured, T_{meas}. The plot in Fig. 2(b) indicates that the biquadratic ordering cannot be suppressed at a temperature higher than the blocking temperature of 60 K. From Figs. 2(b) and 2(d), it is deduced that the blocking temperature is a constant, regardless of the strength of the cooling field.

In order to investigate the stability of the suppression of the biquadratic ordering, we interrupted the hysteresis loop measurements in the first quarter of the hysteresis curve. We were able to verify that the suppression of the biquadratic ordering is stable for at least several days. On the other hand, we found that the suppression of biquadratic ordering is not stable against nonmonotonous variations in the field.

The field-cooling effect may be due to an altered magnetic state of the magnetic moments at the Fe/Cr interface or inside the Cr spacer layer. We used perturbed angular correlation (PAC) spectroscopy to study the field-cooling effect. PAC spectroscopy allows one to probe the magnitude, the direction, and the fraction of the different hyperfine fields.[8] For the experiments, we implanted $(^{111}\mathrm{In})^{111}\mathrm{Cd}$ probes into the sample at 45 keV with a fluence of 10^{13} atoms/cm^2. The spectra were recorded in a high-resolution fast-slow coincidence setup. The sample normal (the [001] axis) and the in-plane [010] easy axis were lying in the plane of the detectors, making an angle of 45° with the detectors. From the measured coincidence spectra, the anisotropy function $R(t)$ is calculated.[11]

The sample was first cooled from 290 K to 15 K in an applied magnetic field of 100 mT along the [010] direction in order to obtain the field-cooling effect. The magnetic field is reduced to 0 mT before the PAC spectrum is recorded. The corresponding anisotropy function is shown in Fig. 3(a). The second PAC spectrum, shown in Fig. 3(b), was recorded after a field sweep—i.e., after cycling the magnetic field to −100 mT, to 100 mT, and back to 0 mT.

The PAC spectra consist of a fast oscillation, due to Cd probes in the Fe layers, and a slow variation, due to Cd probes in the Cr layers. The hyperfine field distribution that corresponds to the best fit to the experimental data is shown in the lower panels of Fig. 3.

The PAC spectra of Fig. 3 display the well-known hyperfine interaction frequency $\nu=93.6(2)$ MHz for ^{111}Cd in Fe at 15 K. Directly after field cooling, only the single frequency

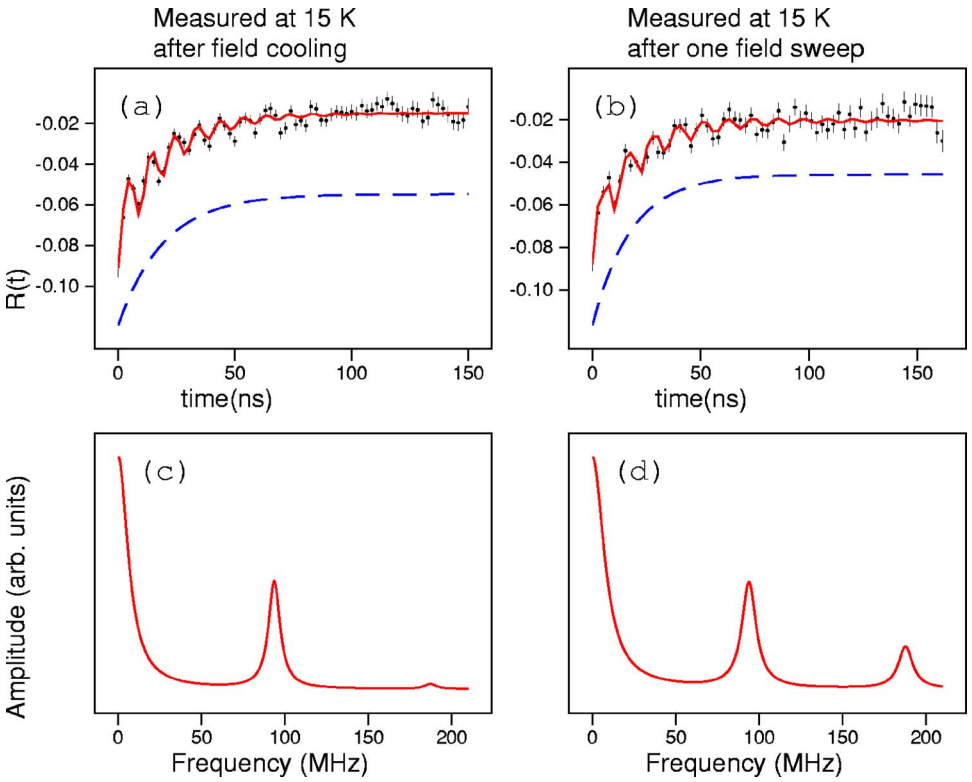

FIG. 3. (Color online) The PAC spectra measured at 15 K after field cooling in 100 mT (a) and after a field sweep (b). The Cr contribution to the spectra is shown separately. The magnetic field was applied along the [010] direction. The hyperfine field distribution of spectra (a) and (b) is shown in panels (c) and (d), respectively.

is observed [Fig. 3(a)], indicating that the Fe magnetization in the two Fe layers is oriented along the field-cooling direction. After a field sweep, both the single and double frequencies are observed [Fig. 3(b)], indicating that the Fe magnetization in the two Fe layers is orthogonally oriented. In other words, the PAC spectra reconfirm the field-cooling effect.

The Cr contribution to the PAC spectra (dashed lines in Fig. 3) consists of a slow decay of the anisotropy, approximated here as a Lorentzian distribution around zero. Remarkably, no difference in the distribution between the suppressed and unsuppressed states of biquadratic ordering is observed. This proves that no major change in the magnetic state of the Cr spacer layer occurs in connection with the field-cooling effect. We therefore conclude that the field-cooling effect is either due to only a subtle change of the magnetic state of the Cr spacer layer or to an altered magnetic state of the magnetic moments at the Fe/Cr interface. Which mechanism is then responsible for the field-cooling effect?

Although the field-cooling effect has certain aspects in common with the *exchange bias* effect,[12] there are also some noticeable differences. First, even at low temperatures, the coercivity of the hysteresis curve for the trilayer system is not markedly different from the one for a single Fe layer grown under similar conditions. Second, the loops are not horizontally shifted. Third, the curve is completely symmetrical after a field sweep. The differing behavior of Fe/Cr/Fe trilayers as compared to conventional exchange-biased systems may be attributed to the specific magnetic properties of Cr thin films. It was demonstrated before that bulklike antiferromagnetic ordering is absent below the critical thickness of 6 nm.[8,9] This is reconfirmed in the present sample as the PAC results described above exclude the presence of spin-density-wave antiferromagnetic ordering either in the zero-field-cooled or field-cooled state. This is also in line with the failure to detect exchange bias effects in Cr/Py bilayers and in Cr/Fe bilayers below a critical Cr thickness of 6 nm.[13,14]

Existing theories on biquadratic interlayer coupling[5] are also not capable of explaining the field-cooling effect. Unlike the thickness-fluctuation mechanism and the magnetic-dipole mechanism, the *loose spin*[15] mechanism is capable of explaining a strong temperature dependence of the biquadratic interlayer coupling. However, in its present form, the loose spin model does not predict an effect of field cooling. It is, nevertheless, conceivable that an extension of the loose spin model in which a portion of the localized moments are allowed to freeze (as considered in Ref. 15) possibly could describe both the temperature dependence of the biquadratic interlayer coupling, on the one hand, *and* the temperature dependence and field dependence of the field-cooling effect on the other hand. Figure 2 may assist theoreticians to verify the appropriateness of a proposed thermodynamic model.

We conclude that it remains unclear which mechanism is responsible for the suppression of the orthogonal ordering at remanence upon field cooling. We hope that the present work will stimulate further experimental and theoretical investigations on this system in the future.

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