Control of Interlayer Exchange Coupling in Fe/Cr/Fe Trilayers by Ion Beam Irradiation

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The manipulation of the antiferromagnetic interlayer coupling in epitaxial Fe/Cr/Fe(001) trilayers by 5 keV He ion beam irradiation has been investigated. It is shown that even for irradiation with low fluences a drastic change in strength of the coupling appears. For thin Cr spacers (below 0.6–0.7 nm) it decreases with fluence, becoming ferromagnetic for fluences above 2×10^{14} ions/cm^2. The effect is connected with the creation of magnetic bridges in the layered system due to atomic exchange events caused by the bombardment. For thicker Cr spacers an enhancement of the antiferromagnetic coupling strength is found. A possible explanation of the enhancement effect is given.

Since the discovery of the antiferromagnetic interlayer exchange coupling effect in the Fe/Cr/Fe layered system by Grünberg *et al.* [1], this effect has been widely investigated both theoretically and experimentally (for a recent review, see [2]). Antiferromagnetically coupled layers are now used in applications such as antiferromagnetically coupled media [3] and artificial antiferromagnets [4]. In many cases such applications greatly gain from a potential of lateral modification of the media parameters with high resolution, after the preparation process of the layered system has been completed. It is not trivial to change the interlayer coupling strength after sample preparation. Until now, to our knowledge, the only reported methods are annealing [5] and charging of the spacer with hydrogen or deuterium [6–8]. However, such techniques can hardly provide any reasonable lateral resolution. On the contrary, beams of light ions with keV energies known for their ability to deeply penetrate into a solid can be focused down to 20 nm [9–11] and should provide a promising pass to accomplish the goal. One of the key advantages of ion irradiation is that magnetic nanopatterning becomes feasible without a change of the sample topography. This is especially important to avoid tribology problems in so-called patterned media [12].

In this Letter, we present first experimental results demonstrating that the strength of the interlayer exchange coupling between two ferromagnets, separated by a nonferromagnetic spacer, can be modified in a controlled manner by ion beam irradiation. It is also shown that for some values of the spacer thickness the ion beam bombardment *enhances* the coupling.

Interaction between two magnetic layers separated by a nonmagnetic spacer layer can be phenomenologically described by:

$$E = -J_1 \cos\phi - J_2 \cos^2\phi$$

where E is the magnetic coupling interface energy, ϕ is the angle between the magnetizations of two magnetic layers, and the parameters J_1 and J_2 represent the strength of the bilinear and biquadratic coupling, respectively [13]. If J_2 dominates and is negative, it promotes perpendicular (90°) orientation of the two magnetization vectors. The microscopic origin of the bilinear coupling is a long-range interaction between the magnetic moments via conduction electrons of the spacer. For smooth interfaces J_1 oscillates as a function of the spacer thickness [14,15]. Essential roughness diminishes the bilinear coupling strength and the amplitude of the oscillations [16,17]. For perfect layered systems J_2 is thought to be small [18]. The experimentally observed strong biquadratic coupling is believed to be due to extrinsic effects [19,20].

Light ion irradiation is known to be an excellent tool to modify magnetic parameters of multilayer systems. Chappert *et al.* [21] have shown that ion irradiation of Co/Pt multilayers leads to a reduction of the perpendicular interface anisotropy. This has been attributed to an interfacial mixing of both atom species. In FePt alloy systems an increase of the perpendicular magnetic anisotropy due to a short-range chemical ordering has been observed after ion irradiation [22]. The technique has been recently applied to exchange-bias systems, consisting of adjacent ferromagnetic and antiferromagnetic layers. It was shown that the magnitude and direction of the exchange-bias field can be tailored by ion irradiation if a magnetic field is applied during bombardment [23,24].

Epitaxial Fe/Cr/Fe(001) samples used in the current studies were prepared in an ultrahigh vacuum molecular-beam epitaxy system with the base pressure below

5×10^{-11} mbar. A Cr buffer with a thickness of 100 nm providing a lattice matched template for the subsequent growth of the Fe/Cr/Fe [25] system was deposited on a MgO(001) substrate. Two Fe films separated by a wedge-shaped, 0.4 to 4 nm thick Cr spacer were deposited on the buffer. Different samples with the thickness of the Fe films from 5 to 10 nm have been prepared. Finally, the system was covered by 3 nm Cr to avoid corrosion for *ex situ* measurements. The details of substrate preparation and the growth procedure are published elsewhere [26]. Figure 1 displays the topography of the lower Fe film and of the Cr spacer as observed by scanning tunneling microscopy (STM). Atomic terraces and monoatomic steps are clearly seen in the images.

Using the measured rms values of the surface roughness (0.11 and 0.18 nm for the Fe and the Cr surface, respectively) and assuming uncorrelated thickness fluctuations for the two films, one obtains an rms value for the thickness fluctuations of the Cr spacer of 0.14 nm, which is close to the thickness of one monolayer (ML). Based on this value and assuming a Gaussian distribution of the probability for the spacer to consist of a given number of monolayers, one obtains, for example, for a nominal thickness of the Cr spacer, d_{Cr} of 4 ML that: 38% of the film area has $d_{Cr} = 4$ ML, 24% has $d_{Cr} = 3$ ML, 6% 2 ML, 0.6% 1 ML, and 0.025% corresponds to direct contact between the two Fe films (so-called "magnetic bridges"). The latter provide a strong ferromagnetic coupling between the Fe films.

Irradiation was performed with 5 keV He$^+$ ions without applied magnetic field with the sample being kept at room temperature. TRIM simulations [27] show that for the used parameter set most ions pass both magnetic layers and are stopped in the Cr buffer layer. The maximum fluence used was 8×10^{14} ions/cm^2. The interlayer coupling was derived from the magnetization curves recorded by longitudinal magneto-optical Kerr-effect magnetometry. A magnetic field of up to 6 kOe was applied in the plane of the sample parallel either to the easy or to the hard magnetic axes of the fourfold magnetic anisotropy of the Fe(001) films.
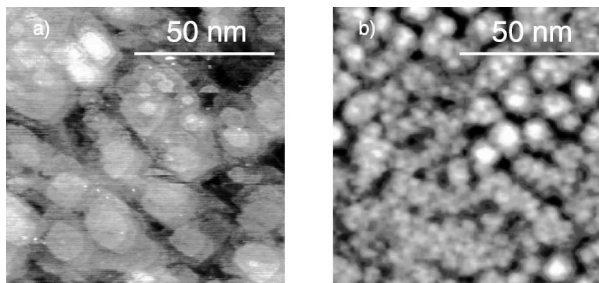
The magnetization curves measured for the field applied along the easy [100] axis show several jumps, characteristic for magnetic double layers with antiferromagnetic and 90° coupling [13]. The saturation field, H_S, which is proportional to $| J_1 + 2J_2 |$ extracted from the magnetization curves is shown in Fig. 2 as a function of the nominal Cr-spacer thickness for different ion irradiation fluences. The data obtained on the as-prepared sample clearly demonstrate both long- and short-period oscillations with a moderate amplitude in agreement with the rms value of the spacer thickness fluctuation obtained from the STM studies. The arrows indicate the first three oscillation maxima of the coupling strength. As is seen in Fig. 2, such a well-prepared layered magnetic system is very sensitive to ion irradiation. The first oscillation maximum ($d_{Cr} = 0.58$ nm = 4 ML) exhibits the strongest effect of the irradiation on the coupling strength. Even the lowest used ion fluence of 0.5×10^{14} ions/cm^2 reduced H_S by nearly 25%. For the fluences above 2×10^{14} ions/cm^2 no antiferromagnetic coupling is observed for this thickness of the Cr spacer. The change of the measured coupling strength for thicker Cr spacers is more intriguing: the coupling *increases* for small ion fluences and then decreases for fluences above 1×10^{14} ions/cm^2.

Of particular interest are the dependencies of the coupling constants J_1 and J_2 as functions of ion fluence and spacer thickness. The constants were derived by fitting the measured remagnetization curves using a standard procedure [2]. The fluence dependence of J_1 and J_2 for the spacer thicknesses corresponding to the first (4 ML), second (6 ML), and third (8 ML) oscillation maxima have been evaluated. Note that only the values of the antiferromagnetic ($J_1 < 0$) and 90°-degree ($J_2 < 0$) coupling constants can be usually derived in such a way. The data is
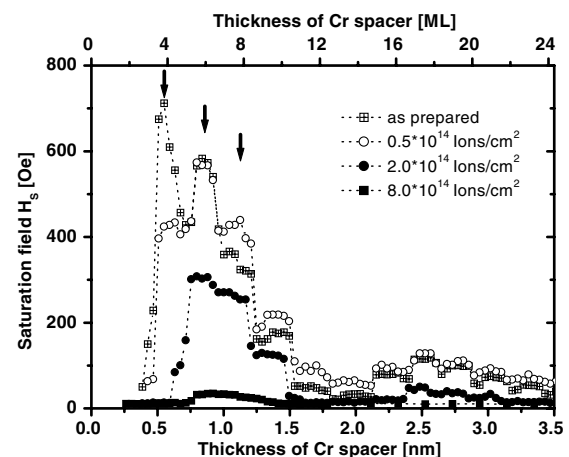

FIG. 1. STM images of the film surfaces: (a) bottom Fe film, *z* scale 1.3 nm, rms = 0.11 nm; (b) Cr spacer, *z* scale 1.5 nm, rms = 0.18 nm.

FIG. 2. Saturation field H_S as a function of the Cr-spacer thickness for the 10 nm Fe/d_{Cr} Cr/10 nm Fe trilayer system as prepared and after the irradiation with the fluences as indicated. The arrows indicate the first three maxima of the interlayer coupling.

presented in Fig. 3. It is clearly seen from the figure that $|J_1|$ strongly decreases with the fluence for $d_{\mathrm{Cr}} = 4$ ML, while it shows a maximum for fluences near 0.5×10^{14} ions/cm^2 for d_{Cr} equal to 6 and 8 ML. $|J_2|$ instead shows a monotonic decrease. Thus, one can conclude from Fig. 3 that the increase of the saturation field at small irradiation fluences is caused by the increase of $|J_1|$.

The origin of the observed phenomena is not understood in all details yet, but they are definitely connected with the surface intermixing caused by the He ions. To understand this qualitatively, let us first consider the fluence dependence of $|J_1|$ for the nominal thickness $d_{\mathrm{Cr}} = 4$ ML. Direct magnetic bridges between the two Fe films provide strong direct ferromagnetic coupling, $J_{\mathrm{direct}} \approx 2A/d_{\mathrm{ML}} = 280$ mJ/m^2, where $A = 2 \times 10^{-11}$ J/m is the bulk exchange constant of Fe and $d_{\mathrm{ML}} = 0.144$ nm is the thickness of one monolayer. The contribution to the measured interlayer coupling due to the magnetic bridges can then be easily estimated, since the percentage area of those bridges is known from the rms analysis of the STM images discussed above [28]. For the as-prepared sample the obtained value of 0.07 mJ/m^2 is essentially smaller than the measured one and indicates that the interlayer coupling via the spacer is an order of magnitude larger than direct coupling across the bridges.

It is known that an ion propagating within a lattice partly dissipates its energy due to nuclear collisions [27]. Such collisions cause recoil of atoms of the lattice and creation of lattice defects and intermixing. Although light ions, such as helium, have a low displacement rate and cause a short range intermixing, these processes are of importance if taking place at the interface. Estimations show that a 5 keV He ion initiates in average between one and two atom pair exchange events per monolayer in Fe and Cr [29]. In the areas with $d_{\mathrm{Cr}} = 1$ ML such an exchange event induces an atomic size

FIG. 3. J_1 and J_2 obtained form the remagnetization curves for different maxima versus the fluence. The lines are guides to the eye.

magnetic bridge. Assuming that each ion generates one exchange event per monolayer as a lower bond and taking into account also a possibility of two successive exchange events at adjacent lattice sites, one obtains that for the fluence of 2×10^{14} ions/cm^2 the relative area of the magnetic bridges increases to 0.2% and their contribution to the interlayer coupling is 0.6 mJ/m^2. This is in rather good agreement with the experimentally observed coupling reduction of 0.72 mJ/m^2. The calculation also demonstrates that the probability for formation of magnetic bridges due to the bombardment decreases exponentially with the nominal spacer thickness at a given interface roughness. Thus, it is not surprising that the effect of the irradiation is weaker for larger spacer thicknesses (the second and third maximum).

Surprising, however, is the observation of an *increase* of the antiferromagnetic coupling strength. These findings might be related to the fact that, first, an intermixing at the Fe/Cr(001) interface with a width of 1–2 ML is supposed to be energetically favorable [30–32], but it is usually not completely achieved during the film growth because of kinetic growth effects. Second, He ions in the discussed energy range very effectively transfer energy to phonons (8–12 eV per monolayer), which in turn help the system to relax into this optimum.

It is known that the interlayer coupling in the Fe/Cr/Fe(001) layered system can be increased by a gentle annealing [5]. Stronger coupling in this case is usually connected with higher lateral homogeneity of interface intermixing between Fe and Cr. On the other hand, a close relation between a homogeneous intermixing at the interface and the interlayer coupling has been recently nicely demonstrated for Fe/Si/Fe [33]: the introduction of two monolayers of $Fe_{0.5}Si_{0.5}$ at every Fe/Si interface brought about a much stronger coupling, as observed on the samples where the intermixing took place naturally.

Using all of the above presented facts, the observed increase of the interlayer coupling can be qualitatively understood as the effect of "phonon annealing": An ion propagating in the lattice creates pulses of hyperthermic phonons along its trajectory. The emitted phonons increase the probability that those parts of the interfaces, where the energetically favorable mixing has not been reached during the growth, move towards this equilibrium. Note here that since this process at its end can produce additional energy, the phonons do not spend their energy and act just as a catalyst. In a similar way as it is observed in the Fe/Si/Fe system [33], a higher degree of the interface homogeneity causes higher interlayer coupling. The proposed model is rather speculative and demands further studies, which are outside the scope of this Letter.

In conclusion, we have experimentally shown that antiferromagnetic interlayer coupling of the Fe/Cr/Fe(001) layered system can be modified using ion beams after
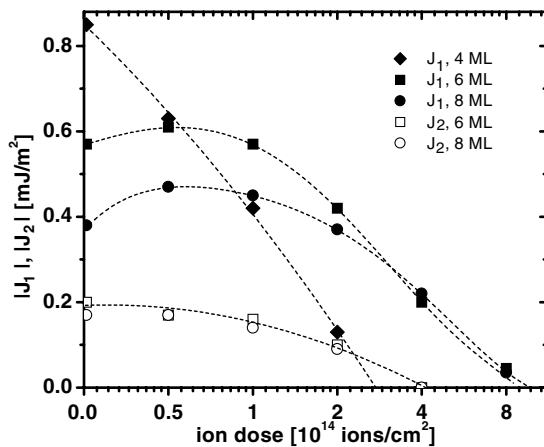
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