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LETTER TO THE EDITOR

Oscillatory exchange coupling in the two-dimensional limit

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Abstract

The dimensionality aspects of the order–disorder transition in single and double magnetic layers is addressed. A single Fe film of three monolayers, embedded in V(001) layers, was determined to be two-dimensional and XY-like. Two Fe layers, separated by 14.4 monolayers of V(001), were determined to belong to the same universality class as the single Fe film. The interlayer exchange coupling was altered *in situ* by introducing hydrogen in the V layers. An oscillation of the ordering temperature was observed, consistent with an oscillatory interlayer exchange coupling. The two-dimensional nature of the bilayer was confirmed by the ratio of the interlayer and intralayer couplings ($|J'/J| \lesssim 2 \times 10^{-3}$). The results therefore support the existence of an oscillatory exchange coupling in the quasi-two-dimensional limit.

The interlayer exchange coupling (IEC) between magnetic layers, separated by a non-magnetic spacer, oscillates in strength and sign as a function of the thickness of the spacer [1]. The reason for the oscillation is the interplay between extension and the shape of the Fermi surface of the spacer [2]. Hence, altering the Fermi surface or the extension of the spacer can be viewed as equivalent routes to influence the IEC.

Large changes in the electronic structure of materials can be accomplished by hydrogen (H) absorption. The presence of H can, for example, induce a metal to insulator transition [3, 4], as well as switch the IEC in metallic superlattices, as shown, for example, in Fe/V(001) superlattices [5]. The influence of the strength of the IEC on the ordering temperature was explored by Leiner *et al* [6, 7], illustrating the possibilities of tuning the inherent magnetic properties of such structures. The possibility of using tailored IEC for exploring the dimensionality aspects of magnetic phase transitions was thereby established.

Close to the critical temperature the behaviour of the magnetization can be approximated by

$$M \propto \left(\frac{T_C - T}{T_C}\right)^{\beta}, \tag{1}$$

where the exponent β is determined by the dimensionality class of the magnetic structure. If the extension of a single magnetic layer is small enough, the magnetic ordering will be two dimensional (2D). The dimensionality of a superlattice made by stacking 2D layers can be altered by the strength of the IEC and the resulting dimensionality can be determined by the deduced exponent β. The resulting dimensionality will be quasi-2D (Q2D) with weak IEC and 3D with strong IEC [8]. Furthermore, when the IEC is changing sign (e.g. from FM (positive) to AFM (negative)), the coupling must cross zero. Hence, the ultimate sign of the presence of weak coupling is the oscillatory exchange coupling (OEC). Therefore the OEC must be accompanied by the presence of Q2D phases, when the individual layers belong to a 2D universality class.

Single Fe films, on various substrates, have previously been shown to be 2D [9–11], and Fe/V superlattices with 2–3 monolayers (ML) of Fe and 7 ML of V have been determined to be 3D [12]. It is therefore tempting to conclude that 2–3 ML Fe layers are 2D with the IEC being responsible for the 3D properties of the aforementioned Fe/V superlattices. However, this could be questioned on grounds of magnetic proximity effects in V [13–15], increasing the effective magnetic thickness of the layers. The dimensionality of a single Fe layer, embedded in V, must therefore be addressed before drawing any general conclusions on the inherent dimensionality of the Fe layers in a multilayered structure. We will therefore start with a discussion on the magnetic properties of a single 3 ML Fe(001) film, embedded in a V sandwich. Thereafter we discuss the influence of H on the magnetic properties of the Fe layer, and the changes in the ordering temperature and magnetization will be addressed. Finally the dimensionality of the magnetic transition of coupled Fe layers will be discussed, and we will demonstrate oscillatory changes in the ordering temperature and the IEC.

The samples were grown on $10 \times 10 \times 0.5$ mm^3 MgO(001) substrates by DC-magnetron sputtering using Ar gas with a purity of 99.9999%, at a background pressure of $\sim 7 \times 10^{-10}$ mbar. The thicknesses of the Fe and the V layers were typically 3 and 14.4 ML respectively. In order to precisely control the thickness of the Fe layers the growth rates for V and Fe were kept low, approximately 0.19 and 0.09 Å s^{-1} respectively. The growth rates were determined from x-ray diffraction measurements on a calibration sample. The samples were capped with a $\sim$30 Å Pd film to prevent oxidation and to improve the uptake of H [16]. The magnetization was determined using the magneto-optical Kerr effect (MOKE); a description of the measurement procedure and setup can be found in [12]. H alloying is accomplished by introducing H_2 gas into the cryostat at room temperature, and the H uptake is monitored by the resistivity of the samples [17][1]. By cooling, further uptake and desorption is effectively hindered, resulting in a constant H concentration at temperatures below 250 K.

Magnetization versus field and temperature of the single layer was measured after exposing the sample to seven different pressures of H_2 gas, in the pressure range 0–4 mbar at room temperature. A selection of these are shown in figure 1. For small H concentrations, $c \propto \sqrt{p}$, which, using previously determined pressure–composition isotherms [18], can be used to estimate the H concentration in the samples. T_C and the exponent β were extracted from fitting a power-law decay convoluted with a Gaussian distribution of T_C. This fitting reproduces the tailing of $M(T)$ above T_C, consistent with a distribution of critical temperatures [19, 12] due to finite-size effects [20]. The results unambiguously prove an increase of the critical temperature with increasing hydrogen content, up to a saturation at approximately 3 mbar.

[1] Although the magnetization is reversible upon H removal, the resistivity is not. Thus we can use the resistivity only as a relative measure of the H concentration. The irreversibility in the resistivity is believed to originate in defect formation in the Pd capping layer.

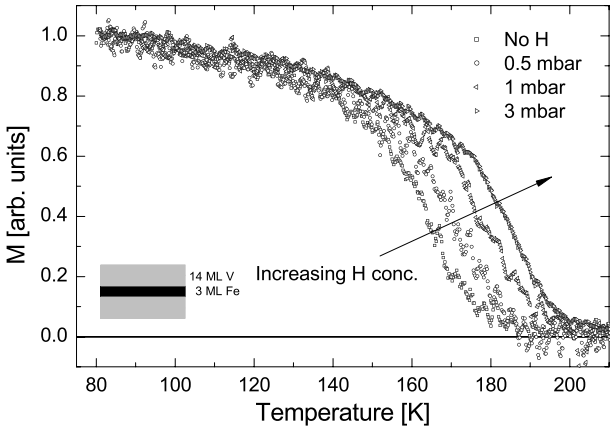

Figure 1. $M(T)$ for the single-layer sample exposed to different H pressures. All datasets are normalized at 80 K. The ordering temperature increases by ∼21 K when the sample is exposed to 3 mbar H at room temperature.

Measurements of the magnetic moments of Fe/V upon H alloying by Labergerie *et al* [16], as well as calculations by Uzdin *et al* [21], give a linear increase[2] of the magnetization with H content. In a homogeneous magnet the intralayer coupling (J) and the magnetization can together be considered as an effective coupling parameter $J_0 = \frac{1}{z}\sum_{k=1}^{z} J_{ik}\langle s_k\rangle$, where z is the number of neighbouring spins coupled to each spin s_i [22]. From this, a linear dependence of T_C with J_0 can be derived. Indeed, we find a linear dependence, $T_C \approx 169.4\ \text{K} + 10.3\sqrt{p}\ \text{K mbar}^{-1/2}$, for $\sqrt{p} \leqslant 2\ \text{mbar}^{1/2}$, which, under the assumption that J is unaffected by the presence of H, verifies the linear increase of the magnetization with H content. Furthermore, the magnetization of 3 ML of Fe embedded in V is consistent with 2D behaviour, and since β is within ±0.04 of the 2D XY value (≈0.23) [20] for all H concentrations the hydrogen-induced changes are not affecting the dimensionality of the magnetic transition. Thus, a single layer of Fe (3 ML) displays 2D behaviour, independent of the H concentration in the V layers.

The critical exponent, β, for strongly coupled layers was determined using an Fe/V sample consisting of three layers of Fe, each 3 ML thick, separated by 7 ML V spacers; see figure 2. If the IEC affects the dimensionality, this should be seen in the temperature dependence of the magnetization. The full line in figure 2 represents the Gaussian fitting of $M(T)$ for this sample, yielding $T_C = 311.2 \pm 0.1$ K and $\beta = 0.358 \pm 0.015$, fully consistent with a 3D transition. The measurements and the analysis of the magnetization data apparently allow the distinction between different dimensionality classes. The proximity of the 2D layers induces a 3D magnetic structure, as expected from the presence of a strong interlayer coupling.

Let us now consider two 2D layers of Fe (3 ML) separated by a V (14.4 ML) layer. The thickness of the magnetic stack in this particular sample is comparable to that of the three-layered magnet discussed above. Hence, if the dimensionality were to be determined by the total extension, the sample would exhibit a 3D transition. Furthermore, if the dimensionality is governed by the IEC, the dimensionality can be either Q2D or 3D, depending on the strength of the IEC. As seen in figure 3, the bilayer exhibits FM behaviour. The analysis of the remanent magnetization, displayed in figure 4, gives $\beta = 0.23 \pm 0.01$. Thus, the magnetization of two 2D Fe layers separated by 14.4 ML of V are indeed 2D XY-like.

[2] In [16] the increase is linear in the range 0–100 mbar.

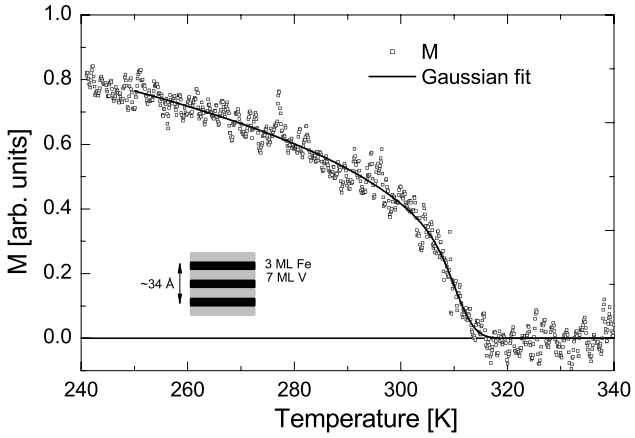

Figure 2. Magnetization versus temperature for a sample with three strongly coupled magnetic layers. The full line represents a fit to the data using a Gaussian distribution of T_C, resulting in $T_C = 311.2$ K and $\beta = 0.358 \pm 0.015$.

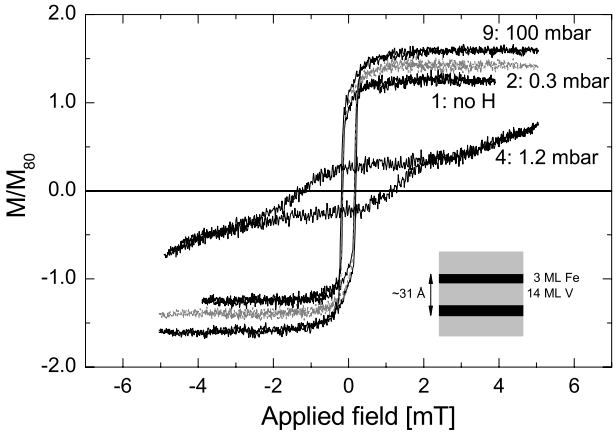

Figure 3. Magnetization versus applied field for the bilayer sample at $T = 0.6\ T_C$ for some pressures of H. The datasets are normalized so that M_{80} is the magnetization of the pristine sample at 80 K. The numbering of the datasets refer to the points in figure 5.

To verify the accuracy of the determination of T_C and β, we employed the linearization method described by Dürr *et al* [9]. The ordering temperature and the exponent β were determined to be 187.4 $\pm$ 0.1 K and 0.230 $\pm$ 0.001 respectively, in the reduced temperature range 0.5–0.98. This confirms the validity of the Gaussian approach in the current context, allowing us to implement an unbiased algorithm for precise determination of the critical temperatures and exponents.

When H is introduced into the sample, the ordering temperature and the remanent magnetization are strongly affected; see figure 4. The ordering temperature versus the square root of the H pressure is shown in figure 5; the inset shows the exponent β versus the square root of the H pressure. A *decrease* in the ordering temperature is observed for the double layer with increasing H content, retaining FM order as seen in figure 3. This decrease must originate in the proximity of the neighbouring Fe layers, as this is opposite to what is observed for a

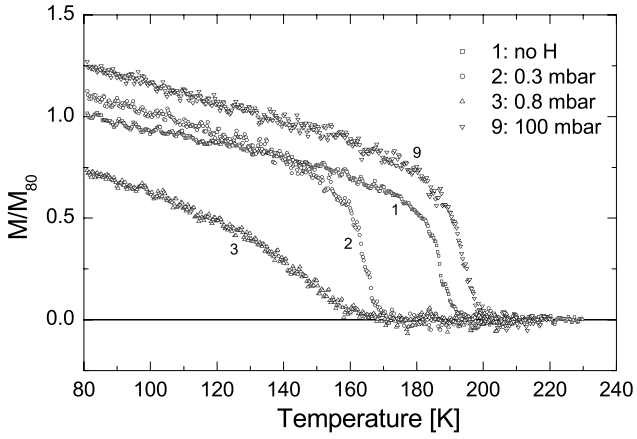

Figure 4. Magnetization versus temperature at different pressures of H for the bilayer sample. All datasets are normalized to the low-temperature value of the measurement on the pristine sample (M_{80}). The numbering of the datasets refers to the points in figure 5.

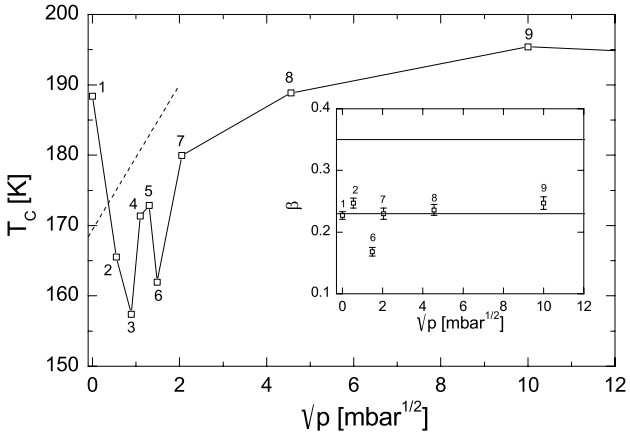

Figure 5. Critical temperature versus $\sqrt{p}$ for the bilayer sample. The dashed line represents the linear increase of T_C as deduced from measurements on the single layer. The uncertainty in T_C is smaller than the size of the symbols. Inset: critical exponent β versus $\sqrt{p}$. The two horizontal lines represent β of the 3D Heisenberg (top) and 2D XY (bottom) models.

single layer. To understand this effect, we therefore need to consider the relation between the ordering temperature and IEC.

In weakly coupled layers, the ratio of the interlayer to intralayer coupling ($\delta = |J'/J|$) can be used to describe the changes of the ordering temperature, in the absence of in-plane anisotropy:

$$T_C \approx T_{C,0} + f(\delta), \tag{2}$$

where $T_{C,0}$ is the critical temperature in the absence of IEC, and where the influence of coupling can be estimated by [20]

$$f(\delta) \approx \frac{K}{(\ln(1/\delta))^2}. \tag{3}$$

Equation (3) describes qualitatively the observed changes: the presence of IEC *always* increases the apparent ordering temperature. The initial decrease in the ordering temperature with increasing H content must therefore originate from a *decrease* of the IEC. The minima in the ordering temperature consequently corresponds to minima in the absolute value of the IEC. Furthermore, for pressures above ∼2 mbar, the IEC is clearly FM, with T_C and magnetization increasing with increasing H pressure. The increase of the ordering temperature can therefore easily be understood as resulting from an increased FM coupling with increasing H content. This is readily confirmed by the well-behaved FM hysteresis loops at high H concentrations.

This leaves only the local maximum unexplained. Recall that the influence of the IEC on the ordering temperature is independent of the sign of the exchange coupling, as seen in (3). This implies that the ordering temperature is independent of the type of ordering. For example, AFM and FM ordered structures would have the same ordering temperature, if the strength of the IEC were the same. As seen in figure 3, the hysteresis loop corresponding to the state close to the local maximum (labelled 4) is not FM-like. A saturation field of 10 mT is estimated by extrapolation, consistent with a negative IEC. This saturation field corresponds to $\delta \approx 1 \times 10^{-5}$, which is extremely weak. However, there is still a remanent contribution at zero field, which is not expected for purely AFM structure. A FM contribution originating from an angle $\neq \pi$ between the magnetic moments of the layers is therefore possible. Hence, the changes cannot be fully reproduced by a linear IEC; a quadratic contribution is required for fully describing the traces of the field dependence of the magnetization.

A semi-quantitative description of the changes in T_C can be deduced by separating the intrinsic effects of H alloying, i.e. the increase of T_C seen in the single layer and the effect on the IEC. The resulting changes in the critical temperature display an oscillatory behaviour similar to that in figure 5 but with the strongest coupling in the pristine state, $\delta \lesssim 2 \times 10^{-3}$ using (2) and (3). With respect to the coupling strength, the sample is expected to be Q2D for all H concentrations.

Now, let us return to the determined exponents of the order–disorder transition. As stated above, the exponent of the virgin sample was determined to be 0.23 ± 0.01, consistent with that of the 2D XY model [20]. This is indeed what is expected for weakly coupled layers. The same conclusion can be drawn from most of the results, as seen in the inset of figure 5. An interesting exception is, however, noted. The point corresponding to the first minimum of T_C exhibits a completely different exponent. This observation does not jeopardize the general conclusion, as this configuration has to belong to a universality class of lower or equal spatial dimensionality than the more strongly coupled layers, as stated above. Thus, the apparent exponent can be the result of competing interactions, resulting in changes in the projected moment at zero field. This is supported by changes in the observed field dependence of the magnetization, at different temperatures. Thus, we conclude that all the exponents corresponding to FM states are indeed Q2D.

In summary, we have shown that a 3 ML Fe film, embedded in V layers, is magnetically two dimensional. On introducing H in the V layers, T_C increases. Since the sample consists of a single magnetic layer, the increase of T_C originates solely in the intrinsic increase of the sample magnetization. The combination of two Fe (3 ML) layers, separated by 14.4 ML of V, exhibits 2D FM behaviour, thus consistent with a weak IEC. By introducing H in the V layers, an oscillation in the ordering temperature is observed. The minima in the ordering temperature define a region with a strong decrease in the remanent magnetization. In this region the IEC <0, with $|J'/J| = \delta \approx 1 \times 10^{-5}$. A confirmation of the dimensionality is thereby obtained, and we establish the existence of an oscillatory interlayer exchange coupling in the quasi-2D limit. These results are conceptually challenging as the changes of the magnetic ordering take place in a dimension which does not contribute to the critical fluctuations.

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