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Absence of dimensional crossover in metallic ferromagnetic superlattices

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Fe/V superlattices are prototypes of atomically thin superstructures. We have measured the ac susceptibility $\chi_{exp}(T)$ for Fe_2/V_5 in the critical regime close to T_C^+. The ordering temperature $T_C=304.75(15)$ K, which is required for an independent analysis of the critical exponent γ in the reduced temperature range below $t=10^{-1}$, is determined separately from the onset of ferromagnetic losses. This analysis yields a $\gamma=1.72(18)$, remaining two-dimensional (2D) Ising-like down to $t=1\times10^{-4}$. In contrast to classical 2D-layered compounds no crossover to three dimensions is observed. This phenomenon in metallic ferromagnetic superlattices is explained by the strong temperature dependence of the interlayer exchange coupling $J'(T)$.

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A classical field to study critical phenomena and dimensional crossover is the investigation of magnetic layered compounds, e.g. $(C_nH_{2n+1}NH_3)_2M\mathrm{Cl}_4$ with $M=$Mn,Cu,Fe and $n=1,2,3,4,10$, where the magnetic ions Mn, Cu, Fe form a single or double layer with an intralayer exchange-coupling constant J and an interlayer exchange-coupling constant J' with $J'/J\ll 1$.[1–4] The magnetic ordering in the plane may be ferromagnetic or antiferromagnetic. In both cases, as one approaches the ordering temperature from the T_C^+ side, one encounters a temperature where the interlayer correlation length $\xi'(T)$ becomes larger than the thickness of the nonmagnetic spacer and the two-dimensional (2D)-layered structure shows a crossover to 3D behavior.[1,3,4] This implies that the magnetic layers are coupled above T_C. In this paper we study metallic ferromagnetic superlattices Fe_2/V_5. The Fe layers are strongly ferromagnetically exchange coupled through V at $T\approx 0$ K ($J_0'\approx 100$ μeV/atom, corresponding to a large effective field of ≈ 50 kOe). Surprisingly, no dimensional crossover occurs: The system remains 2D-like down to 10^{-4} in reduced temperature $t=(T-T_C)/T_C$. This unexpected phenomenon suggests that in metallic superlattices there is a strong temperature dependence of J', which is attributed to temperature-dependent modifications in the Fermi surface of the spacer[5] and the disordering of the magnetic moments with temperature.[6,7] The lack of a dimensional crossover above T_C provides a straightforward evidence that the Fe layers are decoupled at T_C^+. Our work goes beyond interlayer exchange-coupling phenomena and focuses on a clear and systematic analysis to determine T_C and critical exponents in ultrathin-film magnetism.

The $(Fe_2/V_5)_{50}/MgO(001)$ superlattice consists of 2 monolayers (ML) of Fe and 5 ML of V with a repetition rate of 50. From previous works the Fe/V superlattices are known to be of high structural and magnetic homogeneity.[8,9] The magnetic properties of our Fe_2/V_5 samples have been investigated in detail and will be presented elsewhere.[10]

We measured the complex quasistatic susceptibility

$$\chi_{exp}(T)=\chi'_{exp}(T)+i\chi''_{exp}(T) \tag{1}$$

of the Fe_2/V_5 superlattice using a classical mutual inductance (MI) bridge at fixed frequency $f=213$ Hz, calibrated in SI units.[11] Due to the high sensitivity of our MI setup and the large number of repetitions of the Fe_2/V_5 superlattice the use of an extremely small oscillatory field of amplitude $H_0=17$ mOe was possible. This field was applied along the in-plane Fe[110] direction that is the easy axis of magnetization.[10] The use of such small oscillatory fields ($H_0<50$ mOe) is necessary in order to measure the proper initial susceptibility. All external static fields were compensated below 10 mOe by using a pair of calibrated Helmholtz coils. The experiment should be done in thermodynamic equilibrium. Therefore, in this experiment we ramped up and down the temperature with a very slow rate of 5 mK/s to avoid thermal gradients creating artificial curvatures in the susceptibility data, which would result in "effective" critical exponents. The relative error in the temperature determination is ± 50 mK while the absolute error is much larger, but not relevant here.

To determine the critical exponent γ one has to perform a power-law analysis of the internal susceptibility $\chi_{int}(t)$ according to[12]

$$\chi_{int}(t)=\chi_0^+ t^{-\gamma}, \tag{2}$$

where χ_0^+ is the critical amplitude at the T_C^+ side. $\chi_{int}(t)$ is directly related to $\chi_{exp}(t)$:[13]

$$\chi_{exp}^{-1}(t)=\chi_{int}^{-1}(t)+N_{||}. \tag{3}$$

$N_{||}$ is the demagnetizing factor in the [110] plane. While $\chi_{int}(t)$ diverges at T_C according to Eq. (2), $\chi_{exp}(t)$ remains finite due to the demagnetizing field. Consequently, an analysis of $\chi_{exp}(t)$ following the power law of Eq. (2) would then result in an "effective" exponent γ. Therefore, for an accurate analysis of the critical behavior above T_C it is a must to correct $\chi_{exp}(t)$ within Eq. (3) for the demagnetizing effects in order to deduce $\chi_{int}(t)$ and a true γ value.

Equations (2) and (3) include four parameters, namely, T_C, N, χ_0^+, and γ. It is strongly recommended not to perform a four parameter fit, but to determine these values, especially the T_C, separately in order to avoid an interplay of

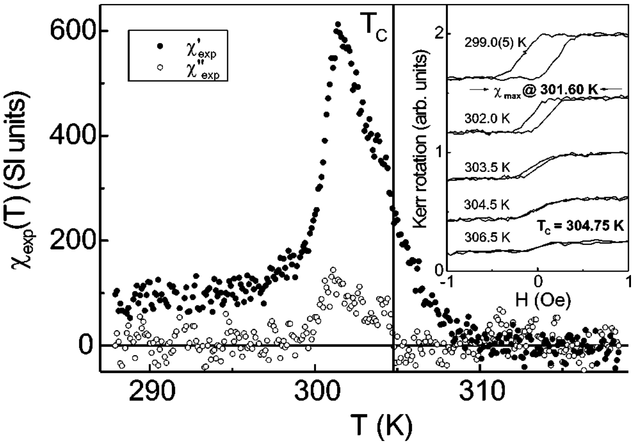

FIG. 1. Real $\chi'_{exp}(T)$ (solid circles) and imaginary part $\chi''_{exp}(T)$ (open circles) of the experimental susceptibility $\chi_{exp}(T)$ at $H_0=17$ mOe. The temperature at the onset of $\chi''_{exp}(T)$ marks $T_C=304.75$ K (vertical solid line). Inset: Temperature-dependent dc-MOKE measurements around $T_{max}=301.6$ K of $\chi'_{exp}(T)$.

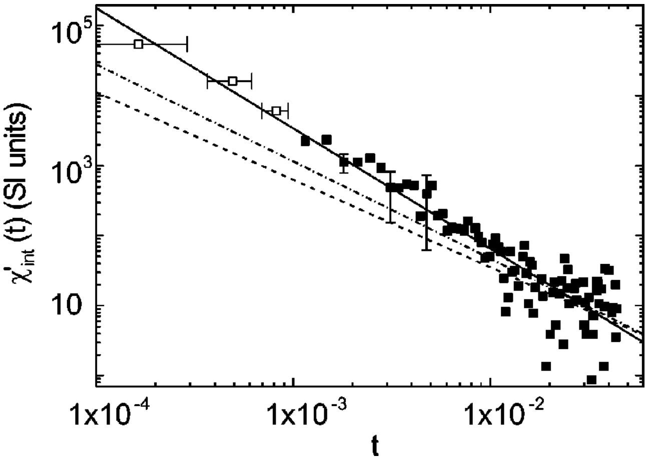

FIG. 2. Double-logarithmic plot of the internal susceptibility $\chi'_{int}(t)$ (open squares indicate a larger error bar). A LSF (solid line) to the data down to $t\approx 1\times 10^{-4}$ yields a 2D-Ising-like critical exponent $\gamma=1.72(18)$. A 3D behavior, both Ising ($\gamma=1.25$, dashed line) as well as Heisenberg ($\gamma=1.387$, dash dotted) is clearly ruled out. The horizontal error bars refer to the relative determination of temperature while the vertical ones to the scattering of the $\chi'_{exp}(T)$ data.

T_C, γ etc. in the fitting procedure.[14] As we do show here, T_C and γ can be also deduced independently from temperature-dependent zero-field ac-susceptibility data. In the following we present a detailed analysis of the ac-susceptibility data discussing the determination of T_C, N, χ_0^+, and γ, respectively.

Figure 1 shows the real (χ'_{exp}) (solid circles) and imaginary parts (χ''_{exp}) (open circles) of the experimental susceptibility χ_{exp} as a function of temperature T. The signal-to-noise ratio of $\chi'_{exp}(T)$ is high (10:1) and the peak full width at half maximum is only ≈ 3 K. This is comparable to the highest quality ultrathin single crystalline metallic films[15,16] and proves the exceptional quality of our Fe_2/V_5 superlattice. As soon as the ordering temperature T_C is reached from the paramagnetic side magnetic energy dissipation occurs. Since the imaginary part of the susceptibility, $\chi''_{exp}(T)$, is a direct measure for magnetic hysteresis losses, one expects the temperature of the onset of $\chi''_{exp}(T)$ to be T_C. Although this is a clear determination of T_C it is not used frequently.[17] The vertical solid line in Fig. 1 marks the onset of $\chi''_{exp}(T)$. Within that independent measure of $\chi''_{exp}(T)$ the ordering temperature is determined to be $T_C=304.75$ K. This value is determined with an accuracy of 0.15 K due to the scattering of $\chi''_{exp}(T)$ data.[18] Clearly, T_C is not located at $\chi'_{exp}(T_{max})$ with $T_{max}=301.6$ K, but about 3 K above. Below T_C, $\chi'_{exp}(T)$ continues to increase due to partial compensation of the demagnetizing field by the anisotropy field that increases gradually in the ferromagnetic phase. In this case an additive term proportional to $(-K/M^2)$ has to be included in the right-hand side of Eq. (3),[19] where K is a function of the anisotropy constants. The maximum of $\chi'_{exp}(T)$ is created by motions of domain walls driven by the oscillatory field. Finally, below T_{max} the coercivity of the sample becomes too large compared to the external oscillatory field and $\chi'_{exp}(T)$ starts to decrease.

That the above given analysis is a correct way to determine T_C (Ref. 20) and to analyze critical phenomena can also be demonstrated with the commonly used magneto-optic Kerr effect (MOKE). Therefore, we carried out longitudinal dc-MOKE measurements at various temperatures around T_{max}. In the inset of Fig. 1 we show MOKE hysteresis curves for temperatures very close to the correct T_C. It is obvious that at T_{max} there cannot be T_C. Considerable hysteresis appears below and still above T_{max} up to $T=303.5$ K, which shows that the system is ferromagnetic. With increasing temperature the hysteresis vanishes completely close to $T=304.5$ K in agreement with the more precise determination via $\chi''_{exp}(T)$. The temperature where the remanence and the coercivity, i.e., hysteresis of in-plane magnetized films vanishes was shown by a combination of MOKE and domain imaging to coincide with T_C.[21] One may understand the vanishing of coercivity at T_C to originate from vanishing magnetization (order parameter) and its anisotropy.

As mentioned above, one needs to determine the demagnetizing factor $N_{||}$ to correct $\chi'_{exp}(T)$ to demagnetizing effects and to calculate the internal susceptibility $\chi_{int}(T)$. Taking into account that the internal susceptibility diverges at T_C, $\chi_{int}(T_C)\rightarrow\infty$,[12] while $\chi'_{exp}(T_C)=233$ remains finite, one obtains via Eq. (3) $N_{||}=1/233=4.3\times 10^{-3}$. This is a reasonable value as we know from previous measurements.[13] After the correction of $\chi'_{exp}(T)$ to the value of $N_{||}$, the internal susceptibility χ'_{int} follows the power law of Eq. (2).

In the next step we determine the critical exponent γ from a double-logarithmic plot of χ'_{int} as a function of t according to Eq. (2). In Fig. 2 the $\chi'_{int}(t)$ data are denoted by solid squares (open squares indicate a larger error bar below $t=10^{-3}$). One may notice that the data indicate a linear behavior down to $t\approx 1\times 10^{-4}$. A least-squares fit (LSF, solid line) yields a critical exponent $\gamma=1.72(18)$ and a critical

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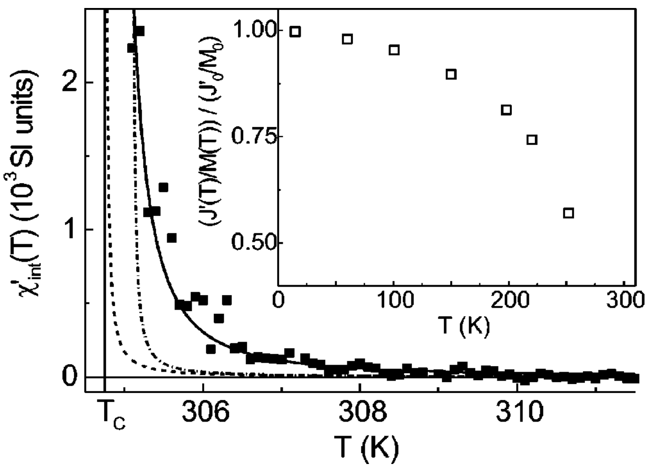

FIG. 3. Internal susceptibility $\chi'_{int}(T)$ (solid squares) and the fit for γ and χ_0^+ from Fig. 2 (solid line). A 3D-Ising $\gamma=1.25$ with T_C fixed does not reproduce the data (dashed line). An additional shift in T_C of 0.35 K fits better (dash-dotted line), but does not reproduce the data at all. Inset: Interlayer exchange coupling $J'(T)/M(T)$ (open squares) normalized to the value at $T=0$ K.

amplitude $\chi_0^+=2.4(6)\times10^{-2}$. The γ value is within the error bar equal to the theoretically predicted value for the 2D Ising model ($\gamma=1.75$).

Baker[22] and Ritchie and Fisher[23] gave an expression for χ_0^+ in Eq. (2) for bcc lattices at T_C:

$$\chi_0^+=\frac{4\pi}{n}\left(\frac{C^+M_0^2}{k_BT_C}\right). \tag{4}$$

Using $C^+\approx1$, the saturation magnetization, T_C, and $n=4.3\times10^{22}/\mathrm{cm}^3$ we calculate a χ_0^+ of the order of 10^{-2}, which is in good agreement with our experimental value of $\chi_0^+=2.4(6)\times10^{-2}$ from Fig. 2.

Since we advocate for a quite rigorous analysis, namely, the separate determination of T_C and γ, one still might question whether another set of T_C and γ parameters is possible. This can be best illustrated in Fig. 3. There we plot χ'_{int} (solid squares) as a linear function of temperature *T*. The solid line is given by the values for $T_C=304.75$ K and χ_0^+ and γ from the LSF in Fig. 2. The vertical solid line indicates the T_C. Now we force ourselves to keep T_C constant and postulate a 3D exponent of $\gamma=1.25$: It can be clearly seen that the dashed line does not meet the susceptibility data at all. On the other hand, we fit the data using a 3D Ising-like γ value and vary T_C slightly. The dash-dotted line is the best possible fit with a shift to higher temperatures of 0.35 K only fitting better the data close to the divergence at T_C. However, it is obvious that the dash-dotted line does not yet reproduce the curvature of the experimental data points.

The classical studies[1,3,4] show that a dimensional crossover occurs at $t\approx10^{-2}$ and the system behaves as 3D by approaching T_C^+. Surprisingly, in our work, there is no change in the slope of $\chi'_{int}(t)$ over the whole reduced temperature range. Even a wrong determination of T_C would not cause a kink in the susceptibility in Fig. 2. This indicates that no dimensional crossover occurs at T_C^+ down to $t\approx1\times10^{-4}$. What can be the reason that this metallic system behaves differently than the numerous examples on the classical ferromagnetic and antiferromagnetic 2D-layered systems?[1–4]

Since in our experiment we do not see a crossover we come to the conclusion that the interlayer exchange interaction itself is strongly temperature dependent and vanishes in the vicinity of T_C. This idea is supported by the inset of Fig. 3. The open symbols show the ratio $J'(T)/M(T)$, $M(T)$ is the temperature-dependent magnetization, normalized to the value J'_0/M_0 at $T=0$ K as it was determined by ferromagnetic resonance (FMR) on the Fe_2/V_5 superlattice below the ordering temperature. Details and a quantitative analysis will be published elsewhere.[10] It is obvious that the $J'(T)/M(T)$ decays strongly close to T_C. Consequently $J'(T)$ itself decays much faster than $M(T)$ at T_C^-. In that sense, the Fe layers are decoupled above T_C and they exhibit a 2D character. One may intuitively expect a dimensional crossover from 2D to 3D below T_C where $J'(T)$ becomes sizable. This matter goes beyond the scope of the present paper that focuses on the T_C^+ side.

A priori there is no straightforward argument why $J'(T)$ should vanish at T_C. From an electronic band structure point of view in a Ruderman-Kittel-Kasuya-Yosida picture an interlayer-exchange-like interaction is mediated by the spin-polarized conduction electrons of the nonferromagnetic spacer, which provide a channel of communication for the magnetic moments of the ferromagnetic layers. Within this framework the temperature dependence of $J'(T)$ is introduced by modifications in the density of states near the Fermi surface.[6] Therefore, considerable reduction of $J'(T)$ is expected only at very large temperatures, close to the Fermi temperature $T_F\approx10^4$ K. Following a similar scenario to the one of the classical layered compounds,[1–4] if $J'(T)$ is present above T_C the magnetic moments are able to ''communicate,'' and as soon as the correlation length exceeds the spacer thickness the superlattice has to behave like a 3D system and a dimensional crossover would be expected at the T_C^+ side. A theoretical scenario that attempts to explain our experimental finding has to focus on the fact that some anomaly happens at $T_C \ll T_F$. Based on the ideas of Ref. 7, we interpret the absence of $J'(T)$ at T_C as an effect of uncorrelated spin-wave excitations on the surfaces of two separated ferromagnetic Fe layers. To our knowledge, up to now there were no published experimental data to evidence for this $J'(T=T_C)$ behavior. The clear experimental finding in the present manuscript is that the Fe/V multilayer (as a prototype system of metallic superlattices) stays 2D down to $t=10^{-4}$ close to T_C.

To our knowledge there is only one other work by Mohan *et al.* providing critical exponents and discussing exchange coupling for metallic multilayers.[25] In that work 1-nm-thick Tb-Dy-Fe magnetic layers were separated by 1-nm-thick Cr spacers and the behavior at T_C was found to be 3D Heisenberg-like. This 3D behavior was attributed to exchange interactions induced by Cr. In this system the interactions through the natural antiferromagnet Cr might be different than through our spacer V. On the other hand the dimensional crossovers above the ordering temperature reported in the classical literature[1,3,4] suggest that the magnetic layers in 2D-layered compounds stay coupled above T_C through various types of interactions, e.g., superexchange interaction,[1] which are of different character than the interlayer exchange coupling of our Fe_2/V_5 sample.

In conclusion, we performed measurements of the temperature-dependent ac susceptibility $\chi_{exp}(T)$ on metallic ferromagnetic superlattices (Fe_2/V_5). After correction to demagnetizing effects a 2D-Ising critical exponent was determined out of $\chi'_{int}(t)$ down to 10^{-4} in the reduced temperature scale. No dimensional crossover to 3D behavior was observed. This is a different phenomenon with respect to all existing literature on classical 2D-layered compounds. FMR measurements show that the interlayer exchange coupling fades away at T_C. This suggests that the critical behavior is determined by the reduced dimensionality of the Fe layers that remain decoupled above the ordering temperature.

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