Size effect on magnetism of Fe thin films in Fe/Ir superlattices

S. Andrieu\textsuperscript{1}, C. Chatelain\textsuperscript{2}, M. Lemine\textsuperscript{1}, B. Berche\textsuperscript{1}, and Ph. Bauer\textsuperscript{3}

\textsuperscript{1}Laboratoire de Physique des Matériaux, Université Henri Poincaré, Nancy 1, BP 239, F-54506 Vandœuvre, France
\textsuperscript{2}Institut für Theoretische Physik, Universität Leipzig, D-04109 Leipzig, Germany
\textsuperscript{3}CREST, Université de Franche-Comté, BP 71427 25211 Montbéliard Cedex, France

(Received November 9, 2018)

In ferromagnetic thin films, the Curie temperature variation with the thickness is always considered as continuous when the thickness is varied from \( n \) to \( n + 1 \) atomic planes. We show that it is not the case for Fe in Fe/Ir superlattices. For an integer number of atomic planes, a unique magnetic transition is observed by susceptibility measurements, whereas two magnetic transitions are observed for fractional numbers of planes. This behavior is attributed to successive transitions of areas with \( n \) and \( n + 1 \) atomic planes, for which the \( T_c \)’s are not the same. Indeed, the magnetic correlation length is presumably shorter than the average size of the terraces. Monte Carlo simulations are performed to support this explanation.

Motivated by both the richness of physical phenomena and the potential applications in magnetoresistive heads, sensors, and more recently spin electronics, there has been considerable progress in the area of thin magnetic films during the last fifteen years. The control of the reduced dimension of the devices, including thin films, multilayers or magnetic dots, became crucial. The progress realized in deposition techniques like sputtering or molecular beam epitaxy (MBE) made possible the deposition of thin films of a thickness of several atomic planes with a very good accuracy and reproducibility. Lithography on the other hand is used to reduce the lateral dimension. This accurate control of the sizes is a necessary condition for preparing model systems in order to investigate, for instance, the magnetic coupling between two magnetic layers via a spacer, the coherent rotation of a single magnetic domain with an applied magnetic field, the spin dependent tunnel current via an oxide, . . . For instance, the well-characterized thin films prepared in UHV conditions can be considered as model systems for studying the variation of the magnetic ordering temperature with the number of deposited atomic planes. This was done for a number of systems - Co, Fe, Ni, FeNi and CoNi on Cu, Gd on (0001) Y, Fe on (001) Ir and Pd. The experimental results were often compared to scaling laws, which predict a universal dependence of the reduced Curie temperature with the number of atomic planes. Power laws are obtained, with universal critical exponents, which depend on the type of magnetic interactions (the so-called universality class). In all previous experimental studies, the Curie temperature was assumed and observed to continuously vary with the quantity of deposited material, even for thicknesses corresponding to a fraction of atomic plane. This is actually true keeping in mind that the magnetic correlation length \( \xi \) diverges at the magnetic transition temperature in the bulk. However, this argument becomes questionable for systems with reduced dimensions. For a fractional number of deposited atomic planes between \( n \) and \( n + 1 \), if the growth occurs layer by layer, the film is made of areas of \( n \) atomic planes while other areas have \( n + 1 \) atomic planes, both areas having separately different ordering temperatures, provided that their lateral extent exceeds the correlation length. If \( \xi \) is lower than the typical size of these areas, it is thus no more justified to assume a unique transition. In this Letter, the observation of two magnetic transitions in a continuous iron film is reported. This effect is actually explained by reduced magnetic coherence length as shown by Monte Carlo simulations.

The preparation and characterization of the Fe/Ir(100) superlattices (SLs) were already reported in previous papers. We thus briefly describe the main properties of iron in these superlattices. The epitaxial growth of Fe on (001) Ir is performed by MBE at 400 K. The Fe growth mode on Ir(100) is Stransky-Krastanov. Indeed, as layer by layer growth takes place up to 5-6 atomic planes, three-dimensional (3D) growth starts above this thickness. Using high-resolution transmission microscopy (HRTEM), we have shown that the SLs constituted of Fe layers up to 6 atomic planes thick exhibit flat interfaces from the first to the last period, whereas interfacial roughness is observed for larger Fe thicknesses, when a 3D growth takes place. Moreover, HRTEM and Mössbauer spectroscopy allow us to control that the Fe structure and magnetic properties are the same for all the Fe layers when no misfit dislocations take place, that is before 3D growth. The structural analyses show that Iron first grows in a body centered tetragonal (bct) structure up to 4 atomic planes. Above this thickness, the additional Fe planes are observed to grow in the bcc structure on top of the bct structure. The control of the thickness is achieved using the RHEED intensity oscillations during the 2D growth. The accuracy on the thickness determination is estimated to be better than 0.1 monolayer (ml). The Fe magnetic properties were investigated using several techniques. First, a Fe mag-
Magnetic ordering is observed for Fe thicknesses above 2 ml, and Ir thicknesses above 3 ml, as shown by SQUID and Mössbauer measurements [8,9]. The Curie temperature varies from 15 K for 3 ml up to 215 K for 6 ml [8]. The magnetization is in-plane and a 2D spin wave behavior is observed for Fe thicknesses up to 6 ml [8]. Low angle neutron scattering shows that the Fe layers are not coupled via Ir for Fe thicknesses up to 6 atomic planes (even down to 4 K). This is an important point, since the order temperature might be affected by this coupling [10]. To summarize, the Fe/Ir SLs may be considered as a set of flat and continuous Fe layers uncoupled with each other. The present work completes this magnetic study, using ac susceptibility ($\chi$) experiments. In order to get a detectable signal, we work on Fe/Ir SLs with 20 periods with Fe thicknesses varying from 4 to 7 planes. The Ir thickness was fixed to 15 Å. The Fe thickness was first accurately determined by fitting the RHEED oscillations. The $\chi$ experiments were first performed on Fe/Ir SLs grown at 400 K with an integer number of Fe atomic planes from 4 to 7, but we also investigated superlattices grown at room temperature in order to check the roughness influence. In the former case, well defined transition peaks are observed (Fig. 1), and the corresponding Curie temperatures are in total agreement with Mössbauer experiments [8].

![Graph of ac susceptibility peaks observed for Fe/Ir multilayers of 20 periods with Ir spacer thickness of 15 Å, and integer numbers of Fe atomic planes.](image1)

Consequently, for an integer number of Fe atomic planes, only one magnetic transition is observed. However, this behavior does not persist for Fe layers consisting of a fraction of atomic planes. In Fig. 3 are shown the $\chi$ spectra obtained on a set of SLs where the Fe thickness is varied from 4 to 5 ml by steps of 0.2 ml. Two peaks are now present, which are approximately located at the transition temperatures observed for 4 and 5 ml, respectively. We observe the same type of behavior for Fe thicknesses between 5 and 6 planes. These results may clearly not be explained by a continuous variation of the Curie temperature with the Fe thickness. Each peak in the $\chi$ spectra arises from a part of the SL with exactly 4 or 5 atomic planes. This could not be explained by some thickness variation on the substrate surface, since in our apparatus the thickness homogeneity is 2% on a 2 inch sample. A thickness dispersion from one Fe layer to the other in the SL could also not be at the origin of these peaks since the incident Fe flux dispersion during the preparation of a SL is better than 5%. We could also imagine that the two magnetic transitions come from the two crystalline bct and bcc phases present above 4 ml. However, this assumption is again not satisfactory. Indeed, for exactly 5 ml of Fe, both bct and bcc phases coexist, but only one transition is observed. This means that both phases have the same $T_c$ if the interface between them is infinite. This is confirmed by Mössbauer spectroscopy, where two different magnetic phases are actually observed, but with the same hyperfine magnetic field [8]. This behavior is also observed on the Co/Ni system for instance [11]. This behavior can finally be explained by the morphology of a Fe growing layer. As the growth is layer by layer, a 4.5 ml thick Fe layer is constituted of some areas with 4 planes and other areas with 5 planes. The magnetic transition observed at 70 K can be attributed to the areas with 4 ml and the other transition at 140 K to the areas with 5 ml. We thus implicitly consider that both areas have their proper Curie temperature despite the fact that they are in contact. This is possible if the magnetic coherence length is smaller than the typical size of the terraces. Consequently, decreasing the distance between the 2D islands in the last atomic plane up to the magnetic coherence length should lead to the occurrence of only one broader peak.

![Graph of ac susceptibility peaks observed for a set of SL with Fe thicknesses varying from 4 to 5 ml (left) and from 5 to 6 ml (right).](image2)
The decrease of the islands separation is possible by decreasing the epitaxial temperature since the dynamical roughness is thus increased. A SL with 4.6 ml per Fe layer was thus grown at room temperature. As shown in Fig. 3, the RHEED patterns obtained at the end of the SL growth are actually typical of a rough surface. The $\chi$ analysis thus shows that, if two peaks are observed for the sample grown at 400 K, only one peak which spreads out is observed for the sample grown at RT, the transition temperature being thus located between the Curie temperatures of 4 and 5 ml samples.

In the following, we report extensive Monte Carlo (MC) simulations which support the previous interpretation of the double-peak structure of the magnetic susceptibility as the signature of the presence of wide terraces in the magnetic layers. These simulations do not intend to reveal the details of the structure of the samples, but are considered as an illustration of the finite-size mechanism which leads to several peaks in the susceptibility signal. For that reason, we have chosen the simplest model of magnet displaying a ferromagnetic-paramagnetic phase transition, the Ising model. We studied qualitatively the effect of a finite lateral length as well as of a finite transverse size of the sample on the susceptibility behavior. These effects are well described in the context of critical phenomena by the standard finite-size scaling theory. Since the Fe layers are magnetically decoupled, and because there is no need of any amplification of the signal in the MC simulations, it is not necessary to pile up layers. We thus considered in the model a layer constituted of two parts with different numbers of atomic planes, let say two regions with $n$ and $n'$ monolayers of magnetic material, which models the juxtaposition of wide terraces having different thicknesses. The boundary conditions are periodic in the plane of the sample and free in the transverse direction. The lattice is a 3D simple cubic lattice, with Ising variables $\sigma_i = 0, 1$ located at the sites of the lattice and interacting via constant nearest neighbor ferromagnetic exchange interactions $J$. In the model, two different magnetic phases with different coupling strengths and different cristallographic structures should a priori be used, but since the Mössbauer hyperfine field is the same, we keep the same $J$ and same structure for both phases in order to get the same $T_c$'s. The Hamiltonian is written $H = -J \sum_{\langle i,j \rangle} \delta_{\sigma_i,\sigma_j}$. This model is the Potts version of an Ising model. In the following, we introduce the variable $K = J/k_B T$. The computation of the thermodynamic properties is performed through MC simulations. Since we are interested in a second-order phase transition, the resort to cluster update algorithms is more convenient, since it is less affected by the critical slowing down than the conventional Metropolis algorithm. In order to avoid time consuming computations at many different temperatures, we also used a histogram reweighting technique.

For the sake of clarity, the main results of the finite-size scaling theory are recalled for consistency. We first consider a simple system of shape $n \times L \times L$. For an ideal system of finite thickness $n$ and infinite lateral extent (thermodynamic limit $L \to \infty$ in the plane), the susceptibility exhibits the usual 2D critical behavior with respect to the deviation $t = |K - K_c(n)|$ from the critical coupling strength $K_c(n)$. $\chi_{\infty}(K) = t^{-\gamma}$. The critical behavior is the same as for a single monolayer, but the critical coupling strength $K_c(n)$ is reduced (or its inverse, the critical temperature, is increased). In the simulations, the systems are of finite size in the three directions. For a fixed thickness $n$ and finite lateral extent $L$, the singularity of the susceptibility expands as $L$ increases, but it remains finite at any temperature. This phenomenon is usually described by a finite-size scaling ansatz $\chi_L(K) = t^{-\gamma} f(L/t^{\nu})$ where $t^{-\nu}$ is the power law behavior of the correlation length of the infinite system and $f(x)$ is a universal scaling function. The susceptibility reaches its maximum for some value of the scaled variable $x^* = L/t_{max}^{\nu}$. The condition of a non singular susceptibility for a finite sample also implies a power law for the scaling function $f(x) \sim x^{\gamma/\nu}$ in the region of the finite-size effects $t^{-\nu} \gg L$ in order to cancel the temperature dependence. The maximum of the susceptibility thus increases as power of the size of the system $\chi_{\max} \sim L^{\gamma/\nu}$, and this mechanism is responsible for the absence of real divergence of $\chi$, as observed in experimental systems where defects are always present which stop the possible divergence of correlations. On the other hand the scaling function simply reaches a constant value far from criticality when $\xi$ is still very small compared to the typical size $L$, in a region where $\chi$ cannot be distinguished from the susceptibility of an infinite system. If $L \to \infty$, $\chi_{\max} \sim L^{\gamma/\nu}$, and this mechanism is responsible for the absence of real divergence of $\chi$, as observed in experimental systems where defects are always present which stop the possible divergence of correlations. On the other hand the scaling function simply reaches a constant value far from criticality when $\xi$ is still very small compared to the typical size $L$, in a region where $\chi$ cannot be distinguished from the susceptibility of an infinite system. If $L \to \infty$, $\chi_{\max} \sim L^{\gamma/\nu}$, and this mechanism is responsible for the absence of real divergence of $\chi$, as observed in experimental systems where defects are always present which stop the possible divergence of correlations. On the other hand the scaling function simply reaches a constant value far from criticality when $\xi$ is still very small compared to the typical size $L$, in a region where $\chi$ cannot be distinguished from the susceptibility of an infinite system.
the behavior is purely 2D, but the thickness \( n \) has also
to be taken into account as a scaling field which induces
a crossover towards a 3D behavior when \( n \gg 1 \). Close
to \( K_c(n) \), a pure 2D critical singularity is thus observed
\( \chi_\infty(K, n) \sim \text{const.} \times t^{-\gamma} \) while a dimensional crossover
is obtained when \( n \gg t_3^{-\nu_{3D}} \) (\( t_{3D} \) is now the deviation
from the bulk critical coupling, \( t_{3D} = |K - K_c(\infty)| \)). The
critical coupling \( K_c(n) \) for example evolves towards the
bulk critical value \( K_c(\infty) \) according to a power law which
involves the 3D correlation length exponent \( \nu_{3D} \).

For a sample with a step between two terraces of \( n \)
and \( n' = n - 1 \) layers and a covering rate which varies
between 0% and 100%, one can observe a susceptibility
with two peaks whose amplitudes strongly depend on
the coverage and whose positions are slightly shifted from
the positions corresponding to pure samples at \( n \) or \( n' \)
monolayers. This is shown in Fig. 4 for \( n = 5 \) for several
covering rates at \( L = 512 \). The two peaks correspond to
two transitions in the thermodynamic limit (this limit is
of course not reached in the real samples where the ter-
racess always have a finite lateral extent). Starting from
the high temperature phase where the system is param-
agnetic, the first peak corresponds to the ordering of the
thicker part of the sample, at a temperature where the
thin part is still disordered. It is a true transition in the
limit \( L \to \infty \) at a given coverage. When the temperature
decreases again, the remaining part of the sample
becomes ferromagnetic and gives rise to a second tran-
sition. The two transitions are not exactly the same:
While the high temperature peak corresponds to an or-
dinary transition, the low temperature transition occurs
in the presence of a ferromagnetic order in the thicker
regions of the sample. For the spins at the interface, in
the vicinity of the step, this is analogous to an extraor-
dinary transition when a surface orders while the bulk is
already ferromagnetic [3].

The scenario proposed here is confirmed by the value of
the correlation length. Outside criticality, the connected
correlation function (after substraction of the square of
the order parameter in the ferromagnetic phase) exhibits
the usual exponential decay \( G_\sigma(r) \sim e^{-r/\xi} \). We can use
this expression to have an evaluation of \( \xi \) at different tem-
peratures. For that purpose, we computed \( G_\sigma(r) \) along
the direction parallel to the step in the plane. An ex-
ponential fit thus leads to the corresponding value of \( \xi \).
Starting from the paramagnetic phase, the correlation
length remains quite small and is not strongly affected
by the presence of the step. As \( K_c(n) \) is increased, \( \xi \)
increases strongly (up to 12 in lattice spacing units) in
the corresponding region, but remains small (\(< 4\)) in
the thin part of the system. We can thus understand
that this first peak in the susceptibility is linked to the
appearance of ordering in the thick regions of the sample,
while the thin part remains paramagnetic. When \( K \)
is increased again, a second singularity develops in \( \chi \),
and is associated to the ordering of this thin part where
\( \xi \approx 6 \). These observations are confirmed by the order parameter
profiles.

To summarize, we have observed that the Curie tem-
perature of Fe in Fe/Ir SL does not vary continuously
with the Fe quantity in the Fe layers. Between 4 and
5 atomic planes, the Fe layer is a mixing of areas with
4 ml and areas with 5 ml, which leads to two magnetic
transitions corresponding to the \( T_c \)'s of 4 and 5 ml. This
behavior is explained by assuming that the lateral mag-
cnetic coherence length is smaller than the distance be-
tween 5 ml thick areas. This assumption is supported
by experiments performed on SLs with rough interfaces.
In that case, only one magnetic transition is observed.
The MC simulations support this explanation. Indeed,
by varying the distance between \( n+1 \) atomic planes thick
areas, the film can exhibit one or two Curie temperatures.
This observation is in fact not surprising, but requires
high quality samples (absence or interface roughness) and
what is more surprising is indeed that such an observa-
tion was never reported in the literature (to the authors
knowledge).

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure4.png}
\caption{Evolution of \( \chi \) as the covering rate is varied be-
tween 4 and 5 ml, 0%, 20%, 40%, 60%, 80%, and 100%
from top to bottom. In the intermediate region, \( \chi \) displays
two peaks located at the “pure” systems temperatures.}
\end{figure}

\[1\] C.M. Schneider, P. Bressler, P. Schuster, J. Kirschner,
J.J. de Miguel, and R. Miranda, Phys. Rev. Lett. 66,
1059 (1990); J. Thomassen, F. May, B. Feldmann, M.
Wuttig, and H. Ibach, Phys. Rev. Lett. 69, 3831 (1992);
B.Ch. Choi, S. Fölsch, M. Farle, and K.H. Rieder, Phys.
Rev. B 56, 3271 (1997); U. Bovensiepen, P. Poupou-
polos, M. Farle, and K. Baberschke, Surf. Sci. 402-404,
396 (1998); F.O. Schumann, S.Z. Wu, G.J. Mankey, and R.F.
Willis, Phys. Rev. B 56, 2668 (1997).

\[2\] C.A. Ballentine, R.L. Fkn, J. Araya-Pochet, and J.L.
Erskine, Phys. Rev. B 41, 2631 (1990); F. Huang, G.J.
Mankey, M.T. Kief, and R.F. Willis, J. Appl. Phys. 73,
6760 (1993).

\[3\] Y. Li and K. Baberschke, Phys. Rev. Lett. 68, 1208
[4] M. Gajdzik, T. Trappmann, C. Srgers, and H.v. Löhneyseen, Phys. Rev. B 57, 3525 (1998).
[5] M. Henkel, S. Andrieu, P. Bauer, and M. Piecuch, Phys. Rev. Lett. 80, 4783 (1998).
[6] H.J. Choi, R.K. Kawakami, E. J. Escorcia-Aparicio, Z.Q. Qiu, J. Pearson, J.S. Jiang, D. Li, and S.D. Bader, Phys. Rev. Lett. 82, 1947 (1999).
[7] S. Andrieu, E. Snoeck, P. Arcade, and M. Piecuch, J. Appl. Phys. 77, 1308 (1995).
[8] S. Andrieu, F. Lahatra-Razafindramisa, E. Snoeck, H. Renevier, A. Barbara, J.M. Tonnerre, M. Brunel, and M. Piecuch, Phys. Rev. B 52, 993 (1995); E. Snoeck, S. Frechengues, M.J. Casanove, C. Roucau, and S. Andrieu, J. Crystal Growth 167, 143 (1996).
[9] O.M. Lemine, Ph.D. Thesis, University of Nancy, 1999, Ph. Bauer, S. Andrieu, O.M. Lemine, and M. Piecuch, J. Magn. Magn. Mater. 165, 220 (1997).
[10] U. Bovensiepen, F. Wilhelm, P. Srivastava, P. Poupopoulos, M. Farle, A. Ney, and K. Baberschke, Phys. Rev. Lett. 81, 2368 (1998).
[11] A. Aspelmeier, M. Tischer, M. Farle, M. Russo, K. Baberschke, and D. Arvanitis, J. Mag. Mag. Mat. 146, 256 (1995).
[12] M.N. Barber in Phase transitions and critical phenomena, vol. 8, C. Domb and J.L. Lebowitz eds. (London: Academic Press, 1983), p.145.
[13] K. Binder in Phase transitions and critical phenomena, vol. 8, C. Domb and J.L. Lebowitz eds. (London: Academic Press, 1983), p.1.