Clarification of contesting results for the total magnetic moment of Ni/Cu(001)

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The aim of this work is to give an experimental answer to the ''contesting results'' concerning the magnetic moments of ultrathin Ni layers. The total magnetic moment of ultrathin Ni/Cu(001) films from 4 to 20 monolayers is measured by an ultrahigh vacuum compatible high-T_c SQUID magnetometer and x-ray magnetic circular dichroism. The magnetization of all Ni films is close to the bulk value for all thicknesses confirming recent theoretical predictions and experiments but contradicts former experimental findings reporting strongly reduced magnetic moments. A reduction of the Ni moment to $0.46\mu_B$/atom is only measurable by capping a 4 monolayer film with Cu. This is dicussed in terms of enhanced surface and reduced interface magnetic moments attributed to hybridization effects between Ni and Cu.

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I. INTRODUCTION

Ni/Cu(001) films have been used during the last decade as a prototype system to study magnetic properties at the ultrathin film limit. A unique spin reorientation transition as a function of film thickness from in to out and then back into the film plane was observed.[1–3] The spin reorientation transition as a function of increasing temperature from in- to out-of-plane was of ''anomalous'' or ''unconventional'' character with respect to other ultrathin film systems.[4,5] This has been explained by the temperature dependence of the various contributions to the magnetic anisotropy energy. While the magnetic anisotropy of Ni/Cu(001) has been intensively investigated and interpreted,[6] a major question concerning the magnitude of the magnetic moments remains unresolved: On the one hand, strongly scattered experimental values were reported (Table I). Namely, reduced magnetic moments of only $0.3\mu_B$/atom compared to the $0.61\mu_B$/atom for bulk were reported for 4 monolayer (ML) Ni/Cu(001).[7] Reduced moments for thicker Cu/Ni/Cu/Si(001) films (17–56 ML) were also reported and attributed to strain effects.[8–10] Recently bulklike magnetic moments of Ni films above 11 ML film thickness[11] and a 10 ML film grown on Co (Ref. 12) have been measured. On the other hand, recent first principles calculations predict almost bulklike moment values for ultrathin Ni/Cu(001) films and a slight reduction of the moment upon Cu capping.[13–16] In this work we deal with the problem of these ''contesting results''[14] for the magnetic moments in ultrathin Ni films. We restricted ourselves to the crucial thickness range between 4 and 20 ML of Ni grown on Cu(001). We show that the Ni moments are indeed larger than previously measured and agree with the theoretically predicted values. Finally, we discuss sources of uncertainty in previous experiments leading to the report of reduced moments.

II. EXPERIMENTAL DETAILS

Ultrathin Ni films between 4 and 9 ML were evaporated through a 3×3 mm^2 mask on Cu(001) as described elsewhere,[17] the thickness determination was done with the help of a quartz balance system. In this thickness range Ni on Cu(001) grows pseudomorphically, fully strained[18] and layer-by-layer.[19] Only a small amount of intermixing occurs which is strictly limited to the interface.[20] The Ni films were magnetically saturated with the help of a pulse-driven electromagnet. Subsequently, the total magnetic moment, i.e., spin plus orbital contribution, is determined with a novel *in situ* UHV-high-T_c SQUID magnetometer[21] at the lowest temperature of 40 K. The magnetic moment per atom can be extracted from the temperature-dependent (40 to 300 K) measurements of the remanent magnetization if the film is in a single domain state, as discussed in Ref. 17. It is a fundamental property of in-plane films that the thermodynamical ground-state is a single domain one.[22] The entire measurement takes only a few minutes, therefore possible contamination due to residual gases is not expected in a base pressure $\leq 2\times10^{-10}$ mbar. Afterwards, the films are capped by about 5 ML of Cu and the process of measurement is repeated once more. That is to say, we did study the effect of Cu capping *in situ* on the same films. To ensure, that our moments deduced from remanent magnetizations are correct, we capped 4 ML Ni with 15 ML of Cu and took it out of the vacuum chamber to measure it via x-ray magnetic circular

TABLE I. ''Contesting'' experimental results for the magnetic moments of thin Ni films. Obviously the findings differ over a wide thickness range.

Ni thickness	Moment (μ_B/atom)
4 ML	0.3(1) (Ref. 7)
4 ML to bulk	0.61(9) This work
11–14 ML	~0.65 (Ref. 11)
3 nm (17 ML)	0.10(9) (Ref. 8)
8 nm (45 ML)	0.23(5) (Ref. 8)
10 nm (56 ML)	0.41(4) (Ref. 8)
15 nm (85 ML)	0.63(3) (Ref. 8)

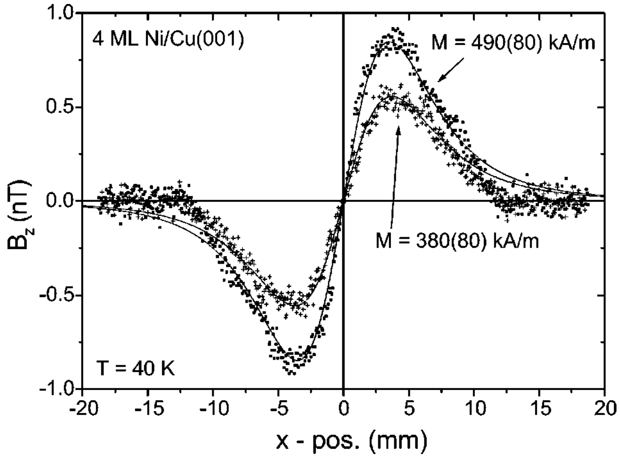

FIG. 1. Measured (data points) and calculated (lines) stray-field distributions for 4 ML Ni/Cu(001) at 40 K before (larger signal) and after Cu capping (smaller signal). A clear reduction of the stray field due to Cu capping is revealed. The fitted magnetizations are given as well.

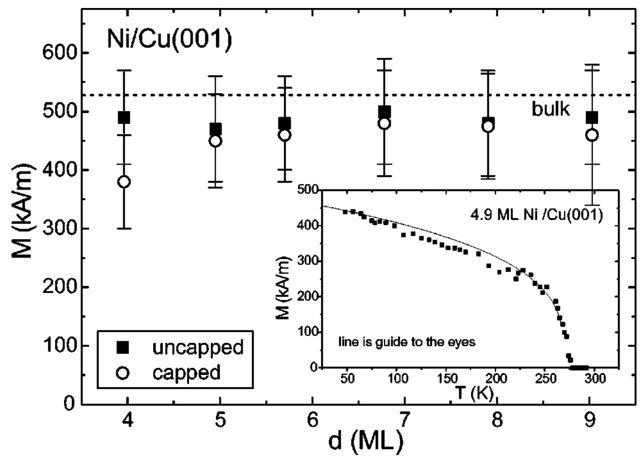

FIG. 2. Measured remanent magnetizations at 40 K for a series of Ni/Cu(001) films between 4 and 9 ML before (solid squares) and after (open circles) Cu capping. All films present bulklike magnetizations except the Cu-capped 4 ML one which shows a decreased magnetization by about 22%. From the temperature dependence of the magnetization shown in the inset one can extract the magnetic moments given in Table I.

dichroism (XMCD) at 10 K and under high magnetic fields up to 3 T to yield magnetic saturation. The XMCD experiments have been performed at the European Synchrotron Radiation Facility (ESRF) in Grenoble (France) at the ID12B beam line[23] at the Ni $L_{2,3}$ edges using the total electron yield detection mode. The degree of polarization of the circular light was 85%. The spectra were recorded either by changing the helicity of the light or by inverting the direction of the magnetic field. A 20 ML thick Ni/Cu(001) capped with Cu and a textured {111} polycrystalline 20 nm thick Ni film sandwiched between Pt were used as references.

III. RESULTS AND DISCUSSION

Figure 1 shows the stray field of a 4 ML Ni/Cu(001) before and after capping with 5 ML of Cu measured with our SQUID magnetometer at a temperature of 40 K. The signal-to-noise ratio for a single measurement is 5/1, in other words the sensitivity of the instrument is better than 1 ML of Ni. By fitting the data according to Ref. 17 we deduce a Ni magnetization of 490(80) kA/m which is close to the bulk value of 528 kA/m. The error is due to uncertainties in determining the distance between SQUID and sample and the thickness calibration since the SQUID is sensitive to the number of contributing magnetic atoms. To verify our calibration thick films have been prepared and they are found to behave bulklike. Measuring 4 ML of Ni at 40 K corresponds to 0.16 –0.19 in a reduced temperature scale assuming a Curie temperature of $T_C=210-250$ K.[6] According to Ref. 17 we extrapolate the temperature-dependent behavior of the magnetization to $T=0$ K and reveal $M(T=0)=524(90)$ kA/m corresponding to $0.61(8)\mu_B$/atom which equals the bulk value. An example for such a measurement is shown in the inset of Fig. 2. Due to the low acessible temperature of 40 K this extrapolation is insensitive to the chosen model for the low-temperature behavior of the magnetization; the $M(T=0)$ values differ by at most 2% for the different models of the T dependence.[17,21] Contrary to previous measurements,[7–10] a reduction of the Ni magnetization by 50% can clearly be ruled out. Subsequent capping by 5 ML of Cu reduces the Ni magnetization by about 22% to 380(80) kA/m compared to the uncapped 4 ML Ni/Cu(001), see Fig. 1. The extrapolation to $T=0$ K yields 410 kA/m which corresponds to $0.47(9)\mu_B$/atom. It should be noted that determining the relative change between the capped and the uncapped films is more significant than providing absolute values of the respective magnetization values as it can be seen from Fig. 1. The reduction of the 4 ML Ni/Cu(001) magnetization by Cu capping confirms recent XMCD studies.[24] More important, it helps to clarify a controversial structural problem. Recent quantitative low energy electron diffraction studies (IV-LEED) claimed the existence of a floating Cu layer on the top of Ni/Cu(001) films.[25] First principles calculations for the interlayer distances of Cu/4 ML Ni/Cu(001) supported this model of a floating Cu layer.[16] However, the magnetic properties are in many cases the most sensitive witnesses of structural changes. Here we show that the large reduction of the Ni magnetization upon Cu capping is inconsistent with pre-existence of Cu on top of the 4 ML Ni/Cu(001) film.

Figure 2 summarizes the results of the remanent magnetizations at $T=40$ K for the full thickness range from 4 to 9 ML Ni/Cu(001) before (solid squares) and after (open circles) capping with 5 ML of Cu. Within the error bar all films possess bulklike magnetizations. Cu capping produces measurable changes of the magnetization only for 4 ML Ni/Cu(001) as discussed above. The measurements are only sensitive to the in-plane [110] component of the magnetization. The constant bulklike value of the remanent magnetization from 9 down to 4 ML of Ni implies that [110] is the easy axis of the magnetization for all films. On the other hand, the reduction of the remanent magnetization for the 4 ML film after Cu capping could originate from a change of the in-plane easy axis from [110] to [100]. Therefore, we measured

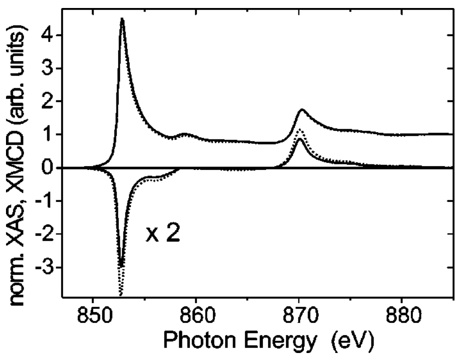

FIG. 3. Normalized x-ray absorption spectra (XAS, linear light) and XMCD (circular light) spectra for Cu/20 ML Ni/Cu(001) (dashed) and Cu/4 ML Ni/Cu(001) (solid line). A clear reduction of the XMCD signal and, consequently, reduction of the magnetic moment is observed by decreasing the Ni thickness yielding $0.44(5)\mu_B$/atom. The measurement was carried out at 10 K under a field of 3 T applied along the film normal (hard axis).

under applied field to clarify the origin of this decrease. In Fig. 3 we show XMCD spectra at the Ni $L_{2,3}$ edges for a Cu/20 ML Ni/Cu(001) and a Cu/4 ML Ni/Cu(001) film. The reduction of the XMCD signal makes it obvious that the Ni moment for 4 ML thickness is decreased. Applying the sum rules[26] and taking the signal of the 20 ML film as reference, we deduce a total Ni moment of $0.44(5)\mu_B$/atom for the 4 ML film. Since there have been previous reports for reduced Ni moments even for 20 ML Ni/Cu(001) (Refs. 8–10) we compared the XMCD spectra of the 20 ML Ni film to the one of a Pt/20 nm Ni/Pt film which was cross checked with high-field SQUID magnetometry.[27] The moment of the 20 ML film equals within 5% the one of the 20 nm Ni film showing that Cu/20 ML Ni/Cu(001) possesses bulklike moments. This conclusion is in full agreement with a recent work by Kuch *et al.* for 11–14 ML Ni/Cu(001).[11]

The value for the magnetic moment of the Cu-capped 4 ML Ni film given by the XMCD is within 6% in agreement with the one that can be determined from the SQUID data if we translate the magnetization value at $T=0$ K into a magnetic moment per atom as discussed above, see Ref. 17. This result indicates that the Ni films measured by the SQUID stay in a single-domain state with the easy-axis along the in-plane [110] for the full thickness range between 4 and 9 ML before and after capping and, therefore, conclusions on the magnetic moments may be safely deduced. In Table II we give the moments for the bulk and 4 ML Ni/Cu(001) from the present experiments, together with the theoretical findings of Refs. 13 and 15. It shows a very good agreement between the present experiment and *first principles* calculations and clearly excludes the possibility that the magnetic moments of Ni films are reduced to an extent as measured before.[7–10] The calculations are carried out for perfect interfaces, while scanning tunneling microscopy has demonstrated that real films have a small amount of intermixing strictly limited to the interfaces.[20] The agreement between the experimental values for real films and theoretical values for ideal films of Table II confirms the results of Ref. 20 that such intermixing has negligible influence on the total magnetic moments of Ni/Cu(001) 4–5 ML thick.

TABLE II. Comparison between the total magnetic moments in μ_B/atom of 4 ML Ni/Cu(001) from the present experiment and previous theoretical works (Refs. 13,15).

	Exp. (this work)	Theory (Ref. 13)	Theory (Ref. 15)
bulk	0.61(8)	0.57[a]	0.72[b] (0.65)
uncapped	0.61(8) (SQUID)	0.52	0.69 (0.62)
capped	0.47(9) (SQUID)		0.61 (0.55)
capped	0.44(5) (XMCD)		
surface	0.88(12)	0.64	0.81 (0.73)
central	0.61(8)	0.55	0.71 (0.64)
interface	0.31(10)	0.37	0.51 (0.46)

[a]In Ref. 13 the moments were calculated in the Korringa-Kohn-Rostoker (KKR) approach and they are assumed to be the spin plus the orbital contribution.

[b]In Ref. 15 spin and orbital moment are calculated by means of the generalized-gradient approximation (GGA). It is known (Ref. 28) that the GGA overestimates the moment by 8–10 %. Therefore, 10% reduced values are given in parenthesis.

In a last step we analyze our experimental data in terms of surface and interface contributions to the magnetic moment, as in Ref. 29. From the moment of the capped films we deduce a reduction of 50% of the Ni moment [$0.31(10)\mu_B$/atom] at the Cu/Ni interface by using the theoretical predictions that reveal bulklike moments for the central Ni layers, which are confirmed by the fact that our experiments show bulklike magnetization for the thicker films. Following the results of theoretical calculations,[13–16,30,31] we attribute this reduction to modifications of the electronic structure of Ni due to the presence of Cu, i.e., to hybridization between Ni and Cu. Separation into surface and interface magnetic moments may be, furthermore, achieved by comparing the capped to the uncapped 4 ML Ni film. Taking into account that the uncapped 4 ML Ni/Cu(001) possesses almost bulk moments, an interface moment of $0.31(10)\mu_B$/atom and $0.61(8)\mu_B$/atom for the central layers, this results in $0.88(12)\mu_B$/atom for the magnetic moment of the surface Ni layer. This large increase is caused by the reduced coordination number of the surface atoms and it is in agreement with the calculations in Refs. 13–16, see Table II. For the Co/Cu(001) system we found that the enhancement of the moment at the surface of $+32(5)\%$ does overcompensate the reduction at the interface which is $-17(3)\%$ leading to an enhancement of the magnetic moment of the entire film.[29] In the case of Ni films on Cu(001) the two contributions just cancel out each other (surface $+50\%$, interface -50%, see Table II). This results in a bulklike magnetic moment for all Ni films between 4 and 9 ML and to the observed reduction of the moment due to Cu-capping, i.e., replacing the surface by a second interface.

The present experiment succeeds in lifting some inconsistency for the Ni magnetic moments between previous experiments and theory. This success is mainly caused by the large year-by-year improvements in magnetometry with monolayer sensitivity, such as our UHV-SQUID magnetometer, and the use of third generation synchrotron radiation facilities. Moreover, the evolution of the ultrahigh vacuum processes, like film growth and structural characterization results in the preparation of higher quality films with lower amounts of defects or gas contamination. For example, compared to Refs. 8,9 the pressure during evaporation of the films was improved by 1 to 2 orders of magnitude which substantially improves the cleanness of the films. Moreover, the interface roughness is drastically reduced in the present experiment (± 1 ML) compared to $\leqslant 10$ Å in Refs. 8,9. The previous findings of reduced moments for the 4 ML Ni films in Ref. 7 based on remanent state measurements along the in-plane [100] direction instead of the in-plane easy axis of magnetization [110]. Accordingly, the projection of M differs by a factor of $\sqrt{2}$. A further slight reduction of the remanent magnetization may originate from a competition between uniaxial step-anisotropies and the small fourfold inplane anisotropy of Ni/Cu(001) films with tetragonal symmetry.

IV. CONCLUSIONS

In conclusion, we presented SQUID and XMCD measurements for the magnetic moments from 4 to 20 ML Ni/Cu(001) ultrathin films before and after capping with Cu. Our results show almost bulklike moments for the full-thickness range and clarify inconsistencies between former experimental work and *first principles* calculations. Having a bulk Ni moment for the central layers, the surface moment of 4 ML Ni is enhanced while the interface moment is reduced by 50% compared to the bulk value. The reduction of the magnetic moment by Cu-capping rules out the possibility of a floating Cu layer on Ni/Cu(001) films.

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[1] B. Schulz and K. Baberschke, Phys. Rev. B **50**, 13 467 (1994).

[2] F. Huang, M. T. Kief, G. J. Mankey, and R. F. Willis, Phys. Rev. B **49**, 3962 (1994).

[3] W. L. O'Brien and B. P. Tonner, J. Appl. Phys. **79**, 5623 (1996); W. L. O'Brien, T. Droubay, and B. P. Tonner, Phys. Rev. B **54**, 9297 (1996).

[4] M. Farle, W. Platow, A. N. Anisimov, P. Poulopoulos, and K. Baberschke, Phys. Rev. B **56**, 5100 (1997).

[5] M. Farle, A. N. Anisimov, W. Platow, P. Poulopoulos, and K. Baberschke, J. Magn. Magn. Mater. **198-199**, 325 (1999).

[6] P. Poulopoulos and K. Baberschke, J. Phys.: Condens. Matter **11**, 9495 (1999).

[7] P. Srivastava, F. Wilhelm, A. Ney, M. Farle, H. Wende, N. Haack, G. Ceballos, and K. Baberschke, Phys. Rev. B **58**, 5701 (1998).

[8] S. Hope, J. Lee, P. Rosenbusch, G. Lauhoff, J. A. C. Bland, A. Ercole, D. Bucknall, J. Penfold, H. J. Lauter, V. Lauter, and R. Cubitt, Phys. Rev. B **55**, 11 422 (1997).

[9] J. Lee, G. Lauhoff, M. Tselepi, S. Hope, P. Rosenbusch, J. A. C. Bland, H. A. Dürr, G. van der Laan, J. Ph. Schillé, and J. A. D. Matthew, Phys. Rev. B **55**, 15 103 (1997).

[10] P. Rosenbusch, J. Lee, G. Lauhoff, and J. A. C. Bland, J. Magn. Magn. Mater. **172**, 19 (1997).

[11] W. Kuch, J. Gilles, S. S. Kang, S. Imada, S. Suga, and J. Kirschner, Phys. Rev. B **62**, 3824 (2000).

[12] S. S. Dhesi, H. A. Dürr, G. van der Laan, E. Dudzik, and N. B. Brookes, Phys. Rev. B **60**, 12 852 (1999).

[13] A. Ernst, M. Lueders, W. M. Temmerman, Z. Szotek, and G. van der Laan, J. Phys.: Condens. Matter **12**, 5599 (2000).

[14] A. Ernst, G. van der Laan, W. M. Temmerman, S. S. Dhesi, and Z. Szotek, Phys. Rev. B **62**, 9543 (2000).

[15] Z. Yang, V. I. Gavrilenko, and Ruqian Wu, Surf. Sci. **447**, 212 (2000).

[16] Z. Yang and Ruqian Wu, Phys. Rev. B **63**, 64 413 (2001).

[17] A. Ney, P. Poulopoulos, M. Farle, and K. Baberschke, Phys. Rev. B **62**, 11 336 (2000).

[18] S. Müller, B. Schulz, G. Kostka, M. Farle, K. Heinz, and K. Baberschke, Surf. Sci. **364**, 235 (1996); W. Platow, U. Bovensiepen, P. Poulopoulos, M. Farle, K. Baberschke, L. Hammer, S. Walter, S. Müller, and K. Heinz, Phys. Rev. B **59**, 12 641 (1999); H. Magnan, P. le Fèvre, A. Midoir, D. Chandesris, H. Jaffrès, A. R. Fert, and J. P. Peyrade, Surf. Sci. **454-456**, 723 (2000).

[19] P. Poulopoulos, J. Lindner, M. Farle, and K. Baberschke, Surf. Sci. **437**, 277 (1999); J. Shen, J. Giergiel, and J. Kirschner, Phys. Rev. B **52**, 8454 (1995).

[20] J. Lindner, P. Poulopoulos, F. Wilhelm, M. Farle, and K. Baberschke, Phys. Rev. B **62**, 10 431 (2000).

[21] A. Ney, Ph.D. thesis, dissertation.de Berlin, 2001, ISBN 3-89825-311-2.

[22] R. Allenspach, J. Magn. Magn. Mater. **129**, 160 (1994).

[23] J. Goulon, N. B. Brookes, C. Gauthier, J. B. Goedkoop, C. Goulon-Ginet, M. Hagelstein, and A. Rogaley, Physica B **208&209**, 199 (1995); N. Drescher, G. Snell, U. Kleineberg, H.-J. Stock, N. Mller, U. Heinzmann, and N. B. Brookes, Rev. Sci. Instrum. **68**, 1939 (1997).

[24] F. Wilhelm, U. Bovensiepen, A. Scherz, P. Poulopoulos, A. Ney, H. Wende, G. Ceballos, and K. Baberschke, J. Magn. Magn. Mater. **222**, 163 (2000).

[25] S. H. Kim, K. S. Lee, H. G. Min, J. Seo, S. C. Hong, T. H. Rho, and J.-S. Kim, Phys. Rev. B **55**, 7904 (1997).

[26] B. T. Thole, P. Carra, F. Sette, and G. van der Laan, Phys. Rev. Lett. **68**, 1943 (1992); P. Carra, B. T. Thole, M. Altarelli, and X. Wang, Phys. Rev. Lett. **70**, 694 (1993).

[27] F. Wilhelm, P. Poulopoulos, G. Ceballos, H. Wende, K. Baberschke, P. Srivastava, D. Benea, H. Ebert, M. Angelakeris, N. K. Flevaris, D. Niarchos, A. Rogalev, and N. B. Brookes, Phys. Rev. Lett. **85**, 413 (2000).

[28] R. Wu (private communication).

[29] A. Ney, P. Poulopoulos, and K. Baberschke, Europhys. Lett. **54**, 820 (2001).

[30] O. Hjortstam, J. Trygg, J. M. Wills, B. Johansson, and O. Eriksson, Phys. Rev. B **53**, 9204 (1996).

[31] A. M. N. Niklasson, B. Johansson, and H. L. Skriver, Phys. Rev. B **59**, 6373 (1999).