Cu cap layer on Ni₈/Cu(001): reorientation and T_C -shift

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Abstract. Adding a non-magnetic Cu overlayer on $Ni_8/Cu(001)$ is known to induce the reorientation of the magnetic easy axis from in-plane to out-of-plane and to reduce the Curie temperature. In this paper both effects are described on the same footing using an effective Heisenberg trilayer. The model takes into account thermal fluctuations of the magnetization and allows to separate explicitly between two possible mechanisms behind the experimental observations, namely between a reduction of the magnetic moment by hybridization and a lattice relaxation at the Ni/Cu interface. Ferromagnetic resonance data for the reorientation and the decreased Curie temperature due to the Cu cap layer are best reproduced by assuming a reduction of the magnetic moment at the interface by $\approx 1/3$.

 $\ensuremath{\textbf{PACS.}}$ 75.10. Jm Quantized spin models – 75.30. Ds Spin waves – 75.70. Ak Magnetic properties of monolayers and thin films

1 Introduction

Thin-film magnetism and related effects have attracted increasing attention of theoreticians and experimentalists alike over the past two decades. The development of molecular beam epitaxy allows to produce ultrathin layers of high quality, and the prospect of further technological applications in the field of nanomagnetism and nanoelectronics ("spintronics") fuels the study of magnetic film structures. Since in general the critical temperature diminishes with decreasing film thickness, one of the key aspects of the theoretical description is to account reliably for the temperature dependence of, e.g., the magnetization.

The decisive role of magnetic anisotropy in systems of reduced translational symmetry has been known for long [1,2]. Magnetic reorientation phenomena are usually governed by different temperature or thickness dependences of the various anisotropies. In the following only second-order contributions to the anisotropic part of the free-energy density are considered [3]:

$$F_a = -\hat{K}_2^t \cos\theta \tag{1}$$

 θ is the equilibrium polar angle between the film normal and the magnetization direction, and any azimuthal anisotropy is neglected. Following the phenomenological model by Gradmann [4] one can write for a film system of thickness d

$$\hat{K}_2^t = \hat{K}_2^V + (\hat{K}_2^{S1} + \hat{K}_2^{S2})/d \tag{2}$$

where $\hat{K}_2^{S\alpha}$ represents the contribution of the film surfaces ($\alpha = 1, 2$) and \hat{K}_2^V is the volume contribution. The temperature-dependence of the different effective contributions to the total anisotropy energy equation (2) can be determined in experiments with varying film thickness, e.g. by ferromagnetic resonance measurements (FMR) [5–7].

For fixed thickness d, the magnetic or spin reorientation transition (SRT) may be driven by temperature. It has been known for long that, if no significantly enhanced surface exchange interaction is present, the magnetization at the surface decreases faster with increasing temperature than within the film [8]. This can be understood as a mean-field effect due the reduced coordination number at the surface as compared to the inner layers. As the layerresolved magnetic anisotropy depends on the corresponding layer-dependent magnetization, the surface anisotropy \hat{K}_2^S decreases faster with increasing temperature than the volume contribution \hat{K}_2^V and a SRT might take place [9].

With respect to the thickness-driven SRT one of the most intensively investigated film systems is Ni/Cu(001). Here, a critical value d_{crit} was found where a transition of the magnetization axis from in-plane to out-of-plane occurs with increasing thickness d [10–12]. When the critical value for pseudomorphic growth is reached, a reorientation back to in-plane takes place. In this work, we focus on a Ni/Cu(001) system just below the first reorientation point. The origin of the thickness-induced transition can be understood as a surface/interface effect: the anisotropies at

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the film surface and in the inner layers of the film favor different directions of the easy magnetic axis – in thin Ni films the surface layers favor the magnetization to align parallel to the film plane whereas the volume anisotropy \hat{K}_2^V tends to an out-of-plane magnetization. In addition dipole-dipole interactions induce a shape anisotropy which favors an in-plane direction of the magnetization of all layers. Since the much stronger ferromagnetic Heisenberg exchange coupling aligns the magnetizations of the individual layers in ultrathin films [13], the surface and volume anisotropies compete to determine the direction of the magnetization. For very thin films, the surface contributions $\hat{K}_2^{S\alpha}$ dominate while for thicker films $d > d_{crit}$, the volume contribution is well-understood today [10].

Another type of reorientation occurs if a cap layer is put on top of Ni/Cu(001) where the thickness of the Ni layer places the system close to the first reorientation from in-plane to out-of-plane described above. This change of the easy direction is the focus of the present work. Without the overlayer, the total anisotropy is dominated by the surface layer contributions. The capping reduces the surface anisotropy so that the volume part of the anisotropy outweighs the surface part, resulting in an easy direction which is parallel to the film normal.

The ferromagnetic resonance technique is an established tool for the evaluation of the anisotropic contributions in thin films [14]. This method probes the uniform spin wave mode $\omega(\mathbf{q} = \mathbf{0})$ of a magnetic sample. An external field **B** is tuned for a given probe frequency $\nu_{hf} = \omega(\mathbf{q} = \mathbf{0})/2\pi$ until resonance occurs at $B_{res}(\theta_B)$, with θ_B being the angle between the magnetic field and the normal to the film plane. The resonance field $B_{res}(\theta_B)$ at which uniform $(\mathbf{q} = \mathbf{0})$ -spin wave modes with the energy $E_{\mathbf{q}=0}^{SW} = h\nu_{hf}$ are excited is the crucial quantity connecting experiment and theory.

One possibility to evaluate the FMR data are the classical Landau-Lifshitz equations [3,14]. At least in their conventional form, however, temperature does not enter explicitly these equations and has to be treated effectively. It has been furthermore pointed out that the behavior suggested by equation (2) is only obtained by considering the total anisotropy at constant reduced temperature T/T_C [15,16]. Since the Curie temperature is thicknessdependent, $T_C = T_C(d)$, this requires the measurement of T_C for any film thickness, implying increased experimental effort and possibly corrupting the experimental data when changing temperature back and forth. The same holds when investigating the effect of a cap layer which also influences the Curie temperature. A theoretical approach which explicitly takes into account the temperature dependence of the magnetization and of the anisotropies appears highly desirable for analyzing FMR measurements.

Recently such an approach based on an extended quantum Heisenberg model was proposed. Thermal excitation of spin waves are accounted for and the temperaturedependence of the anisotropies is obtained in terms of the spin expectation values [17–19]. The theory has been employed to determine the temperature dependence of the magnetic anisotropy [20] in Ni and Co films and of the interlayer exchange coupling in the coupled Ni–Co layered system [21].

In this work we use this approach for investigating the influence of cap layers. As regards non-magnetic overlayers on Ni/Cu(001), Cu is one of the most extensively studied elements [22]. It was found that the critical thickness d_{crit} for the SRT is shifted to lower values $d^*_{crit} < d_{crit}$ when covering the Ni film with Cu [14,23,24], suggesting that the Ni surface anisotropy is reduced by the cap layer. Additionally it was found that the Curie temperature decreases $T_C^* < T_C$ [14,25,26]. Two possible reasons for this have been discussed in literature, namely (i) lattice distortion and (ii) reduction of the magnetic moment at the Ni/Cu interface. In fact up to now it has been found theoretically [27,28] as well as in experiments [6,22,25,29,30] that the latter effect ist mostly responsible for the decrease of the surface anisotropy. The reduction of the magnetic moment of Ni results from the hybridization between the polarized Ni states and the unpolarized Cu states [27]. In [30] it is reported that for four monolayers (ML) Ni on Cu(001), the reduction of the magnetic moment is nearly 50%.

In the next section, our formal treatment of thin magnetic films is discussed [17–21,31]. With the equations given below it is possible to analyze FMR experimental data at any given temperature T. With regard to the present problem of a Cu cap layer on Ni, the hybridization effect is readily accounted for by appropriately choosing the effective spin quantum number of Ni at the interface. Specifically, the resonance fields of FMR measurements for a (un)covered Ni₈/Cu(001) film [5] are fitted. Both the reduction of the magnetic moment at the Ni/Cu interface and the decrease of the Curie temperature due to the capping, described by one and the same approach, are in very good agreement with the experimental findings.

2 Theory

In order to calculate resonance frequencies and resonance fields for comparison with the FMR experiments we employ the Heisenberg model. Such an approach was first proposed in references [17,18]. We use an improved version here including several spin Green functions [19,31,32]. The starting point is the following Hamiltonian:

$$H = -\sum_{\langle ij\rangle\alpha\beta} J_{\alpha\beta} \mathbf{S}_{i\alpha} \mathbf{S}_{j\beta} - \sum_{i\alpha} g\mu_B \mathbf{B} \mathbf{S}_{i\alpha} - \sum_{i\alpha} K_{2\alpha} S_{zi\alpha}^2.$$
(3)

The first term describes nearest-neighbor Heisenberg coupling J between the localized spins $\mathbf{S}_{i\alpha}$ and $\mathbf{S}_{j\beta}$ on the sites i, α and j, β , where Latin indices denote sites within the basal plane and Greek ones indicate film planes (layers). The second term contains an external magnetic field **B** in arbitrary direction with the Landé factor gand the Bohr magneton μ_B . The third term represents second-order lattice anisotropy. $K_{2\alpha}$ are the microscopic anisotropy parameters and $S_{zi\alpha}$ is the z-component of $\mathbf{S}_{i\alpha}$ and is perpendicular to the film plane. The lattice anisotropy favours in-plane ($K_{2\alpha} < 0$) or out-of-plane ($K_{2\alpha} > 0$) orientation. Note that for fixed temperature T the dipolar interaction, which gives an additional easyplane contribution, can be absorbed into the parameter $K_{2\alpha}$ since both have the same angular dependence (cf. Eq. (1)) [19]. Shape anisotropy is therefore not taken exlicitly into account here. Higher-order anisotropy terms are neglected since they are at least two orders of magnitude smaller ($|K_4| \ll |K_2|$) in the considered Ni/Cu(001) system [14,33].

The spin wave excitation spectrum of (3) is evaluated as follows: first the layer-dependent frame is rotated $(\Sigma_{\alpha} \to \Sigma'_{\alpha})$ so that the z'_{α} -axis is parallel to the magnetization in layer α . The equilibrium angles are obtained by requiring the total spin of a layer to be a conserved quantity in the new frame and thus to commute with H':

$$\left[\sum_{i} S_{z'i\alpha}, H'\right]_{-} = 0.$$
⁽⁴⁾

To obtain the above equation spin flips between the layers have been neglected. The equations of motion for the spin Green functions

$$P^{\mu\nu}_{ij\alpha\beta}(E) = \ll S'_{i\alpha\mu}; S'_{j\beta\nu} \gg_E \qquad (\mu,\nu=+,-)$$
(5)

where the +/- refers to the standard spin raising/lowering operator, are solved using the RPA/Tyablikov decoupling [34] and the Anderson-Callen method [35] for the higher Heisenberg exchange and the anisotropy Green functions, respectively. The result can be written after Fourier transformation as follows:

$$\begin{pmatrix} \underline{P}_{\mathbf{q}}^{+-}(E) \ \underline{P}_{\mathbf{q}}^{--}(E) \\ \underline{P}_{\mathbf{q}}^{++}(E) \ \underline{P}_{\mathbf{q}}^{-+}(E) \end{pmatrix} \cdot \begin{bmatrix} \underline{E} - \begin{pmatrix} \underline{M}_{\mathbf{q}}^{+-} \ \underline{M}_{\mathbf{q}}^{++} \\ \underline{M}_{\mathbf{q}}^{--} \ \underline{M}_{\mathbf{q}}^{-+} \end{pmatrix} \end{bmatrix} = \begin{pmatrix} \underline{\eta} & \underline{0} \\ \underline{0} & -\underline{\eta} \end{pmatrix}$$
(6)

The elements of the submatrix building the inhomogeneity matrix on the right-hand side are given by $\eta_{\alpha\beta} \equiv \delta_{\alpha\beta} \langle S_{z'\alpha} \rangle$. Explicitly one obtains the submatrices:

$$M_{\mathbf{q}\alpha\beta}^{+-} = \left(2J_{\mathbf{0}\alpha\alpha}\langle S_{z'\alpha}\rangle + B_{eff}^{\alpha}\right)\delta_{\alpha\beta} \tag{7}$$

$$= -M_{\mathbf{q}\alpha\beta}^{-+} \left(\cos(\theta_{\alpha} - \theta_{\beta}) + 1\right) \langle S_{z'\beta'} \rangle$$
$$= -M_{\mathbf{q}\alpha\beta}^{-+}$$
$$M_{\mathbf{q}\alpha\beta}^{--} = -\frac{1}{2} \sin^{2} \theta_{\alpha} K_{eff}^{\alpha} \delta_{\alpha\beta}$$
(8)
$$= L_{eff} \left(\cos(\theta_{\alpha} - \theta_{\beta}) - 1\right) \langle S_{z'\beta'} \rangle$$

$$-J_{\mathbf{q}\alpha\beta} \left(\cos(\theta_{\alpha} - \theta_{\beta}) - 1\right) \left\langle S_{z'\beta} \right\rangle$$
$$= -M_{\mathbf{q}\alpha\beta}^{++} \tag{9}$$

with the effective field

$$B_{eff}^{\alpha} = 2 \sum_{\gamma}^{\alpha \neq \gamma} J_{0\alpha\gamma} \cos(\theta_{\alpha} - \theta_{\gamma}) \langle S_{z'\gamma} \rangle + B_{x} \sin \theta_{\alpha} + B_{z} \cos\theta_{\alpha} + K_{eff}^{\alpha} (\cos^{2}\theta_{\alpha} - \frac{1}{2} \sin^{2}\theta_{\alpha}), \quad (10)$$

and the effective anisotropy

$$K_{eff}^{\alpha}(T) = 2K_{2\alpha} \langle S_{z'\alpha} \rangle \\ \times \left(1 - \frac{1}{2S_{\alpha}^2} \left[S_{\alpha}(S_{\alpha} + 1) - \langle (S_{z'\alpha})^2 \rangle \right] \right).$$
(11)

Note that the quantity $K_{eff}^{\alpha}(T=0)$ corresponds to the anisotropy field $M_{eff} = 2\hat{K}_2/M - 4\pi M$ (where *M* denotes the total magnetization) in the Landau-Lifshitz theory.

The layer-dependent magnetization $\langle S_{z'\alpha} \rangle$ is calculated self-consistently using a procedure proposed by Callen [36]:

$$\frac{\langle S_{z\prime\alpha}\rangle(T) =}{\frac{(1+\varphi_{\alpha})^{2S_{\alpha}+1}(S_{\alpha}-\varphi_{\alpha})+\varphi_{\alpha}^{2S_{\alpha}+1}(S_{\alpha}+1+\varphi_{\alpha})}{(1+\varphi_{\alpha})^{2S_{\alpha}+1}-\varphi^{2S_{\alpha}+1}} \quad (12)$$

 S_{α} is the layer-dependent spin quantum number and is proportional to the T=0 K-moment in layer α . It will be used below to describe hybridization effects at the Ni/Cu interface. The average magnon occupation number φ_{α} is given by

$$\varphi_{\alpha} = \frac{1}{N} \sum_{\gamma} \sum_{\mathbf{q}} \frac{\chi_{\alpha\gamma}^{+}(\mathbf{q})}{e^{\beta_{T} E_{\gamma}^{+}(\mathbf{q})} - 1} + \frac{\chi_{\alpha\gamma}^{-}(\mathbf{q})}{e^{\beta_{T} E_{\gamma}^{-}(\mathbf{q})} - 1} \,. \tag{13}$$

where the abbreviation $\beta_T = 1/k_B T$ is used. The two terms describe magnon excitations of the system for a given wave vector \mathbf{q} , namely magnon creation ("+") and magnon annihilation ("-"). The excitation energies $E_{\gamma}^{+/-}$ in (13) correspond to the eigenvalues of the supermatrix composed of the $\underline{M}_{\mathbf{q}}^{\mu\nu}$ in (6) and the weights $\chi_{\alpha\gamma}^{+/-}(\mathbf{q})$ are obtained from the eigenvectors in straightforward manner. Equations (4)-(13) represent a closed system of equations which can be solved by iteration.

For a film consisting of η layers there are η magnon branches which are separated by the exchange interaction $\sim J$. Regarding the application of the theory to FMR experimental data, only the lowest spin wave mode is relevant due to the magnitude of the probing frequency ($\sim \nu_{hf} = 9$ GHz). The uniform mode $E_{\mathbf{q}=\mathbf{0}}(B)$ is readily obtained in the course of the numerical evaluation of the above equations.

In order to determine the easy axis of the film, the layer-averaged anisotropy field

$$\overline{K}_{eff} = \sum_{\alpha} w_{\alpha} K_{eff}^{\alpha} \tag{14}$$

is considered. The weighting factor $w_{\alpha} = \langle S_{z'\alpha} \rangle / \sum_{\alpha} \langle S_{z'\alpha} \rangle$ accounts for the layer-dependence of the magnetization which influences the effect of the anisotropy field in a given layer. For $\overline{K}_{eff} > 0$, the easy axis of the system is perpendicular to the film plane whereas for $\overline{K}_{eff} < 0$ the easy axis lies in-plane. In particular, a reduction of the surface anisotropies may lead to a change of sign of \overline{K}_{eff} and thereby induce a SRT for fixed film thickness d.



Fig. 1. Uniform spin wave mode as a function of the external field (left panel) and resonance field as a function of the direction of the external filed (right panel) for positive effective anisotropy $K_{eff} > 0$. The resonance frequency ν_{res} is indicated by the dashed line. Parameters: S = 1, J = 10 meV, T = 0 K, $K_2 = 5\mu_B$ kG, g = 1.



Fig. 2. Same as in Figure 1 but for negative effective anisotropy $(K_2 = -5\mu_B \text{ kG})$.

Figures 1 and 2 show the uniform spin wave mode of a monolayer for T = 0 K as a function of the external field and the corresponding resonance field as a function of the orientation of **B** for positive and for negative effective anisotropy, respectively. For $K_{eff} > 0$ the resonance field is minimal for $\theta_B = 0^\circ (\perp$ to the film plane) and maximal for $\theta_B = 90^\circ (\parallel$ to the film plane). The opposite is found for $K_{eff} < 0$: now the easy direction is parallel to the film plane and the resulting resonance field is minimal for $\theta_B = 90^\circ$ and maximal for $\theta_B = 0^\circ$.

3 Cu cap layer on Ni₈/Cu(001)

For Ni₈/Cu(001) it was found that the easy magnetic axis is parallel to the film plane [14]. Covering the film with 4ML Cu leads to a reorientation, and the Cu₄/Ni₈/Cu(001) film then favors the magnetization to align perpendicular to the film plane. This indicates that



Fig. 3. Cu capping on a Ni film. Only the T = 0 K-moment at the surface is changed $S_{\text{NiCu}} < S_{\text{Ni}}$, the parameters $K_{2\alpha}$, J remain unchanged.

before covering with Cu the surface layers dominate the effective anisotropy \overline{K}_{eff} and thereby the easy magnetic axis. After the capping the contribution of the inner layers, favoring an out-of-plane orientation of the magnetization, dominates the surface part and the easy magnetic axis lies perpendicular to the film plane. As already pointed out, in principle the weakening of the effective anisotropy at the Ni/Cu interface by the capping may be due to two different effects: hybridization or lattice distortion. It appears to be commonly accepted that the dominant mechanism is based on the hybridization of the Ni and Cu states at the interface [6,22,25,27–30]. This is also corroborated by the only minor structural changes in the Ni layer due to the Cu capping [37].

In the following it is demonstrated that using the model presented in the previous section, the assumption of a hybridization-driven decrease of the interface anisotropy is sufficient to quantitatively describe the reorientation of a Ni₈/Cu(001) film in terms of the FMR resonance frequencies [14]. Furthermore, being the new aspect of the present approach to the reorientation, the change in the Curie temperatures before and after the capping is obtained at the same time and is in good agreement with experimental results.

The Ni_8/Cu film is modelled by a trilayer where the outer layers represent the surface layers and the remaining six inner Ni layers are modelled by one center layer. This simplification is justified by the fact that magnetic properties of the subsurface layers are far less affected by the broken translational symmetry than the top and the bottom layer of the Ni slab. The trilayer is sketched in Figure 3 before and after the capping. Assuming that no larger structural changes occur, the microscopic lattice anisotropy parameters are taken to be equal at the Ni/Cu and the vacuum/Ni interface, $K_{2\alpha=1,3} \equiv K_{2sur}$. The exchange interaction parameter $J \equiv J_{\alpha,\beta}$ is fitted to the Curie temperature of the covered film. With a fixed set of parameters $K_{2sur}/K_{2vol}/K_{2sur}$ for the bottom/volume/top layer and J, the FMR resonance fields for the capped and for the uncapped case are fitted by changing only the spin quantum number $S_{\rm Ni} \rightarrow S_{\rm NiCu}$ in the top layer, modelling the change in the magnetic surface moment due to hybridization (see Fig. 3). In order to quantify the magnitude of the surface moment suppression

Table 1. Curie temperature shifts and fit parameters for different ratios of the covered and uncovered T = 0 K-moment.

δ T_C (K) T_C^* (K) ΔT_C (K) $K_{2sur}/K_{2vol}/K_{2sur}$ (μ_B kG) J (meV) $S_{\rm NiCu}$ S_{Ni} 33%425 390 -46.08/32.9/-46.08 4.051 1.5351 2.560% 510387 123-13./4.45/-13.2.47

228

382

we introduce

1

3.5

71%

$$\delta: = 1 - \frac{S_{\rm NiCu}}{S_{\rm Ni}}.$$
 (15)

610

A smaller surface moment leads to a reduced Curie temperature through the reduced effective anisotropy field (11) and via the diminished exchange coupling among the surface spins.

In our calculations we used the same spin quantum number S for the uncovered Ni surface and for the volume layers. One might object that the surface moment of Ni has been reported to be quite enhanced compared to the inner layers [27]. However, a different value of the T = 0 K-moment at the surface would enter our considerations only as an additional fit parameter which would simply change the ratio K_{2sur}/K_{2vol} needed to fit the FMR data. Since no additional information is gained we refrain from this unnecessary complication. We are interested in the relative reduction of the surface moment compared to the interface moment.

Table 1 summarizes the shifts in the Curie temperature and the associated fit parameters for three different ratios of δ . The corresponding resonance frequency curves for the covered (dotted line) and the uncovered (straight line) Ni film at room temperature are shown in Figure 4 together with the experimental FMR results. As can be seen from the different maxima of both spectra, the easy magnetic axis has changed due to the Cu capping from in-plane to out-of-plane direction. In fact all three values of δ yield excellent fits. For $\delta = 1/3$, the results are in good agreement with the measured Curie temperatures of $T_C = 388$ K for the covered and $T_C^* = 444$ K for the uncovered case [33,38]. However, the modification of the surface magnetic moment of the effective 3ML-film overestimates the change of T_C as compared to the case of the real 8MLfilm with a greater number of inner layers. Hence a more realistic value of δ would be a little bit higher than 1/3, in good agreement with the experimentally obtained 50%reduction.

4 Conclusion

In this work we demonstrated the temperature-dependent treatment of a Heisenberg model for investigating different effects caused by capping layers on thin magnetic films. Due to the included spin wave excitations, the theory allows to analyze FMR spectra at any given temperature as well as to evaluate the change of the Curie temperature when increasing the film thickness or adding capping layers. Specifically, we discussed the influence of Cu capping



-7.71/1.9/-7.71

1.6

Fig. 4. The FMR data for the uncapped (capped) (Cu₄)Ni₈/Cu(001) film are taken from reference [14]. Model parameters: $K_{21} = K_{23} = -46.08\mu_B$ kG (surface), $K_{22} = 32.9\mu_B$ kG (interior), J = 4.05 meV, g = 2.1, T = 300 K. $S_{\rm Ni} = 1.5$ for layer interior and $S_{\rm Ni} = 1.5$ resp. $S_{\rm NiCu} = 1$ for the Ni surface resp. Ni/Cu interface.

on a thin $Ni_8/Cu(001)$ film. By assuming a hybridization at the Ni/Cu interface, the experimental FMR results as well as the change of the Curie temperature is captured quantitatively within our approach.

In an extension of the present results, it would be interesting to combine temperature-dependent FMR spectra for varying film thickness with this theory and to investigate other capping layers on thin magnetic films like e.g. Fe or Co films. It is possible to distinguish between different consequences of the capping, namely between lattice distortion (i.e. change in the microscopic anisotropy strength) and hybridization (i.e. reduction of the magnetic moment). Finally, the input parameters can in principle be determined by ab initio calculations. Further research in this direction is planned.

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