Quantum oscillations of properties in magnetic multilayers (invited)

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An oscillatory interlayer exchange coupling observed in many sandwich and multilayer films can be understood as an interference effect of electron waves partially reflected at each interface with spin dependent reflection coefficients. Consequently, we might expect all magnetic properties in some way related to the density of states to oscillate as a function of the magnetic and nonmagnetic layer thickness. In order to experimentally test this concept we have measured different magnetic properties of Ni/Au multilayer films prepared by magnetron sputtering on glass substrates. The Ni thickness was kept constant at $t_{\text{Ni}}=(7.3\pm0.5)$ Å while the Au layer thickness was varied between 4 Å and 80 Å. The films had a coherent fcc structure with (111) texture. The saturation field and the remanence oscillate as a function of $t_{\text{Au}}$ with a period which agrees well with a theoretical value calculated from the bulk Fermi surface of Au and proves that indeed an oscillatory exchange coupling is present. The Curie temperature shows oscillations with $t_{\text{Au}}$ clearly correlated with the exchange coupling constant, $J$: $T_c$ oscillates like the absolute value of $J$. This behavior is indeed expected from mean field theory. Similar oscillations are found for the spin wave parameter and the ground state magnetic moments. The variation of the exchange coupling with temperature and the role of inhomogeneities for the interpretation of the experimental data are discussed. © 1996 American Institute of Physics. [S0021-8979(96)48208-4]

I. INTRODUCTION

Total or partial confinement of itinerant electrons in a solid in one or more dimensions leads to quantized states. These in turn produce a variety of phenomena not observed in bulk material with macroscopic extension. The most widely studied class of material in this respect have been semiconductors: structures fabricated in the form of two-dimensional quantum wells, of quantum wires or quantum dots show a wealth of novel effects in their electronic, optical or transport properties. A simple picture to describe these consequences of electron confinement is the following: at each interface between different materials the moving electron experiences an abrupt change of potential. This is equivalent to a change of the index of refraction for the electron wave causing its partial reflection and partial transmission through the interface. In a layered structure, all the reflected and transmitted electron waves will interfere. Under certain conditions for electron wavelength, layer thickness, and phase shift upon reflection the interference will be constructive or destructive giving rise to oscillations of the density of states as a function of, e.g., layer thickness and in many cases to oscillations of related properties.

The same scenario can be applied to metallic multilayers consisting of ferromagnetic layers separated by nonmagnetic interlayers. An important extension of this simple picture now is brought in by the fact that the (complex) reflection coefficient is different for the two possible spin orientations of the moving electron if the material at one side of the boundary (or both) is magnetically ordered.

In a natural way this general concept leads to a number of phenomena which are to be expected in magnetic layer systems: (i) quantum well states in multilayers and even in nonmagnetic overlayers should be spin polarized; (ii) the total electron spin polarization, i.e., the projected magnetic moment per atom in the (itinerant) ferromagnetic layer should be affected by the layer structure and, in particular, should be an oscillatory function of the nonmagnetic interlayer thickness; (iii) a similar oscillatory behavior is expected for all magnetic properties which are related to the spin dependent density of states in a direct or indirect way like magnetic anisotropy and magneto-optic effects which are both a consequence of spin-orbit coupling; (iv) the total energy of a multilayer system will depend on the relative magnetization directions in adjacent ferromagnetic layers for a given combination of layer thickness, or in other words, the difference of the total energy between parallel and antiparallel alignment will oscillate as a function of layer thickness. This is the well known oscillatory indirect interlayer exchange coupling; (v) other magnetic phenomena which are related to the interlayer exchange coupling directly, e.g., spin wave excitations, or indirectly like the giant magnetoresistance effect (GMR) are also expected to show oscillations with increasing layer thickness.

Historically, the starting point for the study of such quantum oscillations in magnetic multilayers was the discovery first of an antiferromagnetic and later of an oscillatory interlayer exchange coupling in Fe/Cr layered structures. The interlayer exchange and the related GMR effect, which are certainly the most spectacular findings in magnetism during the last decade, have also been the most widely studied subjects in magnetic materials recently. A comprehensive theoretical description of interlayer exchange coupling based on the interference of electron waves has recently been given by Bruno. It is pointed out there that this general concept indeed comprises the different model descriptions proposed in the literature as special cases. A generalization of the concept even allows to explain the exchange coupling across
nonmetallic interlayers. In addition to the well established correlation between the oscillation periods and the Fermi surface of the interlayer material also an oscillatory dependence of the coupling strength on the thickness of the ferromagnetic layer is expected in this framework; this indeed was found subsequently in several experiments.\(^5\)\(^6\)\(^7\)

In the literature a large number of experimental and theoretical studies can be found on topics (iv) and (v) mentioned above; most of them are focused on the influence of layer thickness and structure, interface sharpness, chemical composition, etc. on interlayer coupling and magnetoresistance. However, relatively few results have been reported concerning the other predictions stated above. The existence of quantum well states with spin dependent density of states in Cu overlayers on Co and Ag on Fe has been demonstrated by photoemission and inverse photoemission experiments.\(^7\)\(^\)\(^8\)\(^9\) Recently, oscillations of the spin wave parameter, \(\sigma\), with an interlayer coupling constant \(J_1\) of the ferromagnetic layers \(M\) were found.

An indication of an oscillating magnetic anisotropy of Co ultrathin films successively covered with a nonmagnetic metal (Au, Cu, Pd, etc.) of increasing thickness has been seen in several experiments.\(^10\)\(^–\)\(^12\) Recently, oscillations of the Kerr rotation angle from an ultrathin Co film as a function of the thickness of a Au overlayer have been observed and related to spin polarized quantum well states in the Au.\(^13\) On the other hand, oscillations of ground state magnetic moments and the Curie temperature have not been reported until now.

Within the category of magnetic properties directly related to interlayer exchange coupling, the shape of the magnetization curve is by far the most widely used criterion for the presence of this indirect coupling: low remanence is often interpreted as an indication of antiferromagnetic coupling and the saturation field can directly provide the coupling strength in the antiferromagnetic case. Brillouin light scattering and FMR are also affected by interlayer coupling and can be used to evaluate the coupling strength.

Thermal spin wave excitations are closely connected with exchange coupling between spins. Within the Heisenberg model of exchange interaction between nearest neighbors the spin wave stiffness constant is proportional to the (average) exchange integral and, therefore, should be modified by the presence of an interlayer exchange coupling. Such an effect has indeed been observed in Fe/Ag multilayer films\(^14\) where an oscillation of the spin wave parameter, \(B\), with Ag layer thickness was found.

In an analogous way, the Curie temperature, \(T_C\), of a ferromagnet is proportional to the average exchange energy per atom within mean field theory (MFT). Consequently, we might expect the Curie temperature of a periodic stack of ferromagnetic and nonmagnetic layers to oscillate as a function of the thickness of the nonmagnetic layer. This can be seen if we assume the component of the total exchange energy density due to interlayer coupling, \(E_{\text{inter}}\), to be given by

\[
E_{\text{ex}}^{\text{inter}} = -J_1 \cdot \hat{M}_a \cdot \hat{M}_b
\]

with an interlayer coupling constant \(J_1\) oscillating between positive and negative values depending on the nonmagnetic layer thickness \(t\).

A change in sign of \(J_1\) will reverse the relative alignment of the magnetizations \(\hat{M}_a, \hat{M}_b\) of the ferromagnetic layers from parallel to antiparallel or vice versa in the equilibrium state. So, \(E_{\text{ex}}^{\text{inter}}\) is always negative and depends only on the absolute value of \(J_1\). If we now assume the Curie temperature to be proportional to the total exchange energy per atom which is the sum of interlayer and intralayer coupling energy, then we expect \(T_C\) to oscillate as a function of the interlayer thickness with a period of oscillation half of the period of \(J_1\). However, no experimental verification of this effect has been reported in the literature previously.

To check the general validity of the concept outlined above we have investigated several magnetic properties of a series of Ni/Au multilayer films. Ni was chosen because of its relatively low Curie temperature; by sufficiently reducing the Ni layer thickness the Curie temperature can be reduced below room temperature. This allows to eliminate the risk of irreversible structural changes when measuring around the critical point. For the nonmagnetic layer Au was used because it can easily be grown with a strong (111) texture on glass substrates even by sputtering. The magnetization curves showed oscillations of the remanence and the saturation field with the period of the Au layer thickness theoretically predicted for the interlayer exchange coupling through AuNi(111), \(J_1\). The Curie temperature showed pronounced oscillations with a period like \(J_1\) in accordance with the mean field argument given above.\(^15\) Oscillations of the spin wave parameter were observed which are clearly correlated with the oscillations of \(T_C\). A similar behavior was found for the ground state magnetic moments.

II. EXPERIMENT

For sample preparation dc magnetron sputtering sources were used in an UHV based sputtering system. The argon pressure during deposition was 6\( \times \)\(10^{-3}\) mbar, the sputtering rate was 0.27 nm/s for Au and 0.13 nm/s for Ni.

To study the interlayer coupling a series of Au(30 nm)/[Ni(\(t\))Au(\(t\))]/Au(30 nm) multilayers \((t_{\text{Ni}}=0.73\pm 0.05 \text{ nm}, t_{\text{Au}}=0.4 – 8.0 \text{ nm})\) was deposited on glass substrates at room temperature. A second series of samples was prepared with \(t_{\text{Ni}}=0.4 – 1.4 \text{ nm}\) and \(t_{\text{Au}}=8.0 \text{ nm}\) ("uncoupled films") in order to determine the dependence of \(T_C\) on the thickness of the Ni layers alone. The Au layer is expected to be thick enough to exclude any interlayer exchange coupling. The data of this series were used to correct for the effect of the variation of \(t_{\text{Ni}}\) (\(\pm 0.05 \text{ nm}\)) on \(T_C\) within the first series (see below).

The total amount of Ni and Au of each sample was determined by x-ray fluorescence analysis (XFA) and converted into a nominal layer thickness assuming the bulk densities. The statistical error is \(\pm 0.006 \text{ nm}\) for Ni and \(\pm 0.016 \text{ nm}\) for Au. The variation of the Ni thickness within the first series of \(\pm 0.05 \text{ nm}\) was due to the finite precision of the shutter operation.

Small angle x-ray diffraction and high resolution electron microscopy (HREM) were used to study the structure of the films. The layer thickness or multilayer period determined in this way agreed with the XFA data with an error below 1\%.

The films are polycrystalline with a pronounced (111) texture. For \(t_{\text{Au}}=2 \text{ nm}\) a coherent fcc structure is observed

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FIG. 1. Saturation field, $H_{sat}^{exp}$, and magnetic remanence, $m_R/m_S$, versus Au layer thickness, $t_{Au}$, at 10 K. Dashed lines mark the behavior expected without interlayer coupling. Solid lines represent a spline fit and serve as a guide for the eye.

plained by a certain uncorrelated interface roughness which creates local thickness fluctuations. The presence of such a roughness can also be concluded from x-ray diffraction and HREM data. As a consequence, interface regions with ferromagnetically coupled ones giving rise to a finite remanence.

From the interlayer coupling contribution to the saturation field, $H_{sat}^{exp}$ (Fig. 1), we can estimate the interlayer coupling energy per unit area, $I_1$: if we assume that for $H = H_{sat} = 2H_{sat}^{exp}$ the interlayer exchange energy is balanced by the magnetostatic energy of the Ni layers in the applied field we get $I_1 \approx 0.004$ erg cm$^{-2}$ for the first antiferromagnetic maximum at $t_{Au} = 1.2$ nm.

FIG. 2. Determination of the Curie temperature, $T_C$, by two different methods: (a) linear extrapolation of $m^2(T)$ to $m=0$; (b) minimum of $dm/dT$.

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III. EXCHANGE COUPLING

Like in previous studies magnetization curves of the films were used to detect the presence of an interlayer exchange coupling. The magnetic field was always applied parallel to the film plane since all samples exhibit a magnetic easy plane anisotropy. The spontaneous magnetic moment, $m_S$, was determined by linear extrapolation of the saturated part of the magnetization loop between 2 and 10 kOe back to zero field. The remanent moment, $m_R$, is the magnetic moment in zero field after saturation at 50 kOe. $H_{sat}^{exp}$ is taken as the external magnetic field for which the sample moment reaches 0.8 $m_S$, 80% saturation has been chosen for convenience because this value can be determined with much better precision than the real saturation field $H_{sat}$. The latter is approximately twice the value of $H_{sat}^{exp}$ for all samples.

The plot of the saturation field, $H_{sat}^{exp}$, and the magnetic remanence, $m_R/m_S$, at 10 K as a function of Au layers thickness, $t_{Au}$, (Fig. 1) clearly shows an oscillatory interlayer coupling. The observed oscillation period, $\Lambda_{exp} = (1.15 \pm 0.1)$ nm, is in good agreement with the value $\Lambda_{theor} = 1.135$ nm theoretically predicted on the basis of experimental data on the Fermi surface of bulk gold.\footnote{1} The additional continuous increase of the saturation field with increasing Au layer thickness is probably due to local magnetic anisotropies caused by the increasing strain in the Ni layers. It is also observed that $m_R/m_S$ does not reach zero at its minima which correspond to the strongest antiferromagnetic coupling. This can be explained by a certain uncorrelated interface roughness which creates local thickness fluctuations. The presence of such a roughness can also be concluded from x-ray diffraction and HREM data. As a consequence, interface regions with ferromagnetically coupled ones giving rise to a finite remanence.

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IV. CURIE TEMPERATURE

The Curie temperature was determined from the magnetic moment vs temperature $m(T)$ measured with the SQUID magnetometer. The data were taken at decreasing temperature in the presence of a constant magnetic field of 100 Oe parallel to the film plane. This approach will also work in the case of antiferromagnetic coupling provided that the moments do not completely cancel; this does not happen in practice. From $m(T)$ curves $T_C$ is evaluated by two different methods as shown in Fig. 2: either by linear extrapolation of $m^2(T)$ to $m=0$ as suggested by molecular field theory [Fig. 2(a)] (fitting with $m^2 = (T_C - T)/T_C$) with $T_C$ and $\beta$ as free parameters yields similar results but with less precision) or by the minimum of $dm/dT$ [Fig. 2(b)]. The first value is expected to be closer to the true transition temperature than the second one which, however, can be determined with higher accuracy. Both values are shifted by a constant amount of $(5 \pm 0.5)$ K relative to each other for all films. Therefore, the difference is irrelevant for the question of a possible variation of $T_C$ with the Au layer thickness. $T_C$ values discussed below were determined according to Fig. 2(b).

The dependence of the Curie temperature on the thickness of the Ni layers, $t_{Ni}$, has been determined using the uncoupled films mentioned above. The results are shown in Fig. 3. In order to interpolate between the experimental data points, the following power law is used:

$$T_C(t_{Ni}) = T_C(\infty) \cdot \left[ 1 - \left( t_{Ni}/t_0 \right)^{-\lambda} \right].$$

\footnote{1}
suggested by scaling theory. A numerical fit to the experimental data yields $\lambda = 0.63 \pm 0.02$ and $t_0 = (0.422 \pm 0.009) \text{ nm}$ while $T_C(\infty) = 630 \text{ K}$ was held constant. These values are in excellent agreement with those found by Childress et al. As already mentioned above, within the first series of films with a nominal Ni thickness of 0.73 nm a certain variation of the Ni layer thickness could not be avoided ($\pm 0.05 \text{ nm}$). In order to eliminate the effect of the Ni layer thickness on $T_C$ the data of Fig. 3 were used. It was assumed that the variation of $T_C$ with $t_{Ni}$, $dT_C/dt_{Ni}$, is the same for all multilayers. This would not be exactly true for films with thin Au layers if $T_C$ is indeed affected by interlayer exchange. However, for all films the intralayer exchange energy is still much larger than the interlayer exchange energy and, therefore, the procedure used will be approximately correct. This point will be discussed further at the end of this section.

The corrected Curie temperatures, $T_C(t_{Au})$, of all samples from the first series are plotted in Fig. 4(a) in comparison with the magnetic remanence, $m_d/m_s(t_{Au})$ [Fig. 4(b)], as a function of Au layer thickness. It is apparent that $T_C$ does not increase monotonically with decreasing thickness of the Au interlayer but rather shows pronounced oscillations. There is an unambiguous correlation between the Curie temperature, $T_C$, and the remanence, $m_d/m_s$: maxima of $T_C$ are observed for those values of $t_{Au}$ where $m_d/m_s$ has a maximum or a minimum; minima of $T_C$ occur wherever the interlayer exchange coupling is zero, i.e., when $m_d/m_s$ equals the value observed for very thick Au interlayers. It means that the Curie temperature of the Ni layers is enhanced both by a ferromagnetic and an antiferromagnetic interlayer coupling. This is exactly what is expected from the argument given above that the interlayer exchange energy has always the same sign both for ferro- and antiferromagnetic interlayer coupling. Furthermore, this type of oscillation with a shorter period compared to the oscillation of $J_1$ supports the conclusion that the observed $T_C$ oscillation is not an artefact produced by the method used to determine $T_C$ because otherwise we might expect the same oscillation period for $m_d$ (i.e., $J_1$) and $T_C$.

We can also rule out a possible interpretation of the oscillatory Curie temperature as a consequence of some structural changes with increasing Au layer thickness by the following arguments: (1) if a structural change affects $T_C$ it is hard to understand how this could cause an oscillation of $T_C$; (2) the close correlation between the oscillations of $T_C$ and of the remanence and saturation field is a strong indication of a common origin of both phenomena; (3) the observed oscillation period agrees well with the value which was theoretically predicted on the basis of the Au Fermi surface; (4) oscillations of the spin wave parameter, $B$, and the magnetic ground-state moments of Ni have been found from magnetization measurements at temperatures far below $T_C$ which are strongly correlated with the $T_C$ oscillations. These will be discussed in the next section.

Mean field theory predicts the critical temperature of a ferromagnet to be related to the exchange energy per magnetic atom, $e_{ex}$, according to $k_B T_C = e_{ex}$ for an fcc lattice with $S=1$. This allows us to estimate the interlayer coupling energy per atom from the variation of the Curie temperature, $\Delta T_C = 40 \text{ K}$, between $t_{Au} = 1.2 \text{ nm}$ and $t_{Au} = 1.3 \text{ nm}$ resulting in $\Delta T_C = 40 \text{ K}$, $\Delta T_C = 3.4 \text{ meV}$.

For the calculation of the coupling energy per unit area, $I_1$, we need the areal density of those Ni atoms which are responsible for the enhancement of $T_C$ due to interlayer exchange coupling. The simplest assumption is that all Ni atoms are equally affected by interlayer coupling; however, it might be more realistic to assume that only a certain part of
the Ni atoms experience the maximum coupling strength which determines $T_C$. Using the first assumption we obtain a [coupling energy density of $I_1=36$ erg/cm$^2$. This value seems unrealistically large, i.e., several orders of magnitude larger than the coupling strength derived from the saturation field (0.004 erg/cm$^2$) and also much larger than theoretical values\(^4\) (e.g., 0.4 erg/cm$^2$).

On the other hand, we know from the measurement of the remanence discussed above that there is no homogeneous antiferromagnetic coupling in our multilayer films. In this case local values of $e_{ex}$ will have a broad distribution and the Curie temperature will be determined rather by the maximum of $e_{ex}$ than by its average value. Therefore, $I_1$ will be drastically smaller than the first estimate. A more elaborate theory of phase transitions in exchange coupled multilayer films should also consider the crossover from a 2-dimensional to a 3-dimensional phase transition when the interlayer coupling is switched on.

A final remark must be made to the procedure applied above to correct for the variation of the Ni layer thickness within the series of coupled multilayers. After it has been shown that $T_C$ is indeed affected by interlayer exchange it is also clear that Eq. (2) cannot hold in general for exchange coupled multilayers because the coupling strength is known to depend also on the ferromagnetic layer thickness.\(^3\)\(^,\)\(^5\)\(^,\)\(^6\) Once this dependence is measured for Ni/Au multilayers a complete discussion of the Curie temperature as a function of $t_{Ni}$ and $t_{Au}$ will be possible for the coupled films.

V. SPIN WAVES AND GROUND STATE MAGNETIC MOMENTS

The spontaneous magnetic moment of the coupled multilayer films, $m_s$, was determined from the magnetization loops by extrapolation to $H=0$ as described above. The temperature dependence $m_s(T)$ was fitted by two different expressions which have been proposed in the literature for spin wave excitations in thin films, i.e.,

$$m_s(T) = m_s(0) \cdot (1 - B \cdot T^{3/2})$$

and

$$m_s(T) = m_s(0) \cdot (1 - A_0 \cdot T \cdot \ln(1 + T)).$$

The validity of spin wave theory being limited to sufficiently low temperature, the discussion was restricted to $T<T_C/2$.

Within the experimental uncertainty the $T^{3/2}$ law valid in bulk ferromagnets gave an equally good fit to the data as a $T \ln T$ law which was suggested in the literature for monolayer films. The approximate validity of an “effective $T^{3/2}$ law” for surfaces and ultrathin films has been pointed out before.\(^21\) Therefore, both the parameter $B$ from the $T^{3/2}$ fit and the interlayer coupling constant, $J_1$, derived from a $T \ln T$ fit according to Qiu et al.\(^20\) have been calculated. It turned out that a $T \ln T$ fit yields a very large uncertainty of $J_1$; therefore no meaningful values of $J_1$ could be obtained and no clear correlation with the exchange coupling strength calculated from the saturation field (Sec. III) was found. Hence, we exclusively consider the effective $T^{3/2}$ law in the following. From the corresponding fit to the data we obtain the spin wave parameter $B$ and the average ground state magnetic moment $\langle \mu \rangle$.

It is well known\(^21\) that the spin wave parameter $B$ increases with decreasing thickness of a ferromagnetic film. This can be understood in the following way: From spin wave theory on the basis of the Heisenberg model we know that $B$ is proportional to $D_0^{-3/2}$ and the spinwave stiffness constant, $D_0$, is proportional to the exchange energy per atom. In an inhomogeneous system we assume $D_0$ to scale with the average exchange energy per magnetic atom; hence, in an ultrathin film due to the reduced coordination at the interfaces $D_0$ will be reduced and $B$ enhanced. This argument would therefore predict an enhancement of $D_0$ by an interlayer exchange coupling of any sign, i.e., a decrease of the spin wave parameter.

In order to study the influence of the interlayer coupling on the parameter $B$ we have to eliminate the influence of thickness variations of the Ni layer within the series of “coupled films” like it was done for the Curie temperature in Sec. IV. We follow the same principle by first measuring $B(t_{Ni})$ for the uncoupled films and use a numerical interpolation to calculate $B(t_{Au}, t_{Ni}=0.73 \text{ nm})$ for the coupled films. With the same reasoning as before we assume that the variation of $B$ with $t_{Ni}$, $dB/dt_{Ni}$, is the same for both series of samples.

The corrected values of the spin wave parameter are plotted versus Au layer thickness in Fig. 4(c) in comparison to the remanence as a measure of the interlayer coupling [Fig. 4(b)] and to the Curie temperature [Fig. 4(a)]. Qualitatively, the correlation of $B$ to $m_R$ is the same as for $T_C$: minima of the spin wave parameter, i.e., maxima of spin wave stiffness coincide with strongest ferro- (=$ maximum of $m_R$) or antiferromagnetic (=minimum of $m_R$) coupling, maxima of $B$ occur for zero interlayer exchange (e.g., for $t_{Au}=1 \text{ nm}$ and 1.3 nm). This means that both ferromagnetic and antiferromagnetic interlayer coupling stabilize the magnetic order in the multilayer stack against thermal fluctuations.

The same qualitative result was obtained by Keavney et al.\(^14\) for Fe/Ag multilayer films. They determined the $B$ parameter (or more precisely, a prefactor, $k$, which accounts for the interlayer coupling) from the interface hyperfine field in relatively thick Fe layers (>20 ML). Using their numerical analysis\(^14\) we are able to estimate the interlayer coupling constant, $J_1$, from the variation amplitude of $B$ and get values around 0.5 erg/cm$^2$ for the largest coupling energy density. This result agrees with the value given by theoretical estimates.\(^4\)

Finally we discuss the ground state magnetic moments obtained from the fits to $m_s(T)$ with a $T^{3/2}$ law: by dividing the total saturation moment of the sample for $T=0$ by the total number of Ni atoms which are directly obtained by XFA we determine the average ground state atomic moment ($\langle \mu \rangle$).

The result is shown in Fig. 4(d) in comparison to the other quantities discussed above. Clear variations are seen and the dependence of $\langle \mu \rangle$ on $t_{Au}$ qualitatively follows the Curie temperature. It is worth mentioning that both quantities have been determined by completely independent measurements in different temperature regimes. Apparently, a ferromagnetic or antiferromagnetic interlayer coupling not only stabilizes the magnetic order at finite temperatures but also enhances
the ground state moments in the itinerant ferromagnet compared to the uncoupled ultrathin layer. The effect of interlayer coupling on ground state moments has been studied theoretically using a tight binding scheme for the Fe/Cr system\textsuperscript{22} and indeed a correlation between the coupling and the magnetic moments was found. For Ni/Au, however, no calculations of the moments are known and, in particular, first principles calculations are lacking.

The average Ni moment is considerably reduced compared to bulk Ni ($\mu = 0.6\mu_B$). This may be a consequence of hybridization of Ni states at the interfaces. A more detailed discussion of this effect in connection with band calculations will be presented elsewhere.

VI. DISCUSSION AND CONCLUSIONS

It has been shown for Ni(111)/Au(111) multilayered films that all the magnetic properties investigated here—remanence, saturation field, Curie temperature, spin wave stiffness and ground state magnetic moments—show pronounced oscillations as a function of the Au interlayer thickness between 3 and 12 ML. These observations confirm the basic idea that in a metallic multilayer film consisting of ferromagnetic layers separated by nonmagnetic interlayers all magnetic properties should reflect the interference of electron waves which are produced by multiple spin-dependent reflections at the interfaces.

The oscillations observed for the different quantities agree in their wavelength and their phase within the uncertainty of the present experiments. The coupling strength determined from the different properties, however, differ substantially. Several circumstances might account for this fact:

(i) The respective properties are measured at quite different temperatures ranging from $T = 0$ up to $T_C$. The interlayer coupling constant itself is known to vary with temperature; this has been found experimentally\textsuperscript{23} and studied theoretically from the standpoint of the Fermi–Dirac statistics of the electrons\textsuperscript{24} as well as in relation to spin wave interactions;\textsuperscript{25} however, no data are available in the literature which refer to the Ni/Au system.

In particular, our data show that the interlayer exchange coupling does not vanish when the critical temperature of the interlayer ferromagnetic order is approached. This is not \textit{a priori} evident and merits further theoretical work.

If $J_1$ varies with temperature then the spin wave parameter, $B$, of a coupled multilayer will also depend on temperature. This is not compatible with the established methods of measuring $B$ because they assume $B$ to be constant in a finite temperature range. For a quantitative comparison with more elaborate theories a different fitting scheme will have to be employed.

(ii) In a strongly simplified model we have assumed that all the oscillatory quantities are uniform within each layer in our films. This is not realistic; instead, it is certain that neither the ground state moments nor the spin deviation by spin waves and the interlayer exchange coupling are homogenized in the perpendicular direction within the individual Ni layers. We indeed measure average values by our magnetometric technique as pointed out earlier. In addition, we have to expect lateral inhomogeneities like different grain orientations, layer thickness fluctuations and roughness. We indeed measure average values by our magnetometric technique as pointed out earlier. In addition, we have to expect lateral inhomogeneities like different grain orientations, layer thickness fluctuations and roughness. It would be helpful to measure depth profiles of the magnetic moments and spin wave parameter by using Mössbauer spectroscopy with probe layers. However, this method is practically restricted to Fe and not applicable to Ni; $T_C$ of Fe, on the other hand, is so high that its variation with interlayer coupling could only be studied on superlattices with 1 monolayer Fe films between Au layers.

As a consequence of the arguments given above similar experiments will be carried out on epitaxially grown films with controlled flatness and better structural uniformity. It is expected that this will allow a more quantitative study of the phenomena discussed in this communication, in particular the oscillations of the Curie temperature observed for the first time. Complementary efforts will be made to use a more comprehensive theory of thermal spin waves and phase transitions as well as band calculations for the ground state magnetic moments in exchange coupled multilayer films.

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