A drop of hyperfine field at Sn in Fe/Cr/Sn/Cr multilayers

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The magnetism of the Fe/Cr/Sn/Cr multilayers was studied by first-principles functional theory. The calculations by LAPW and SKKR methods showed that two solutions exist in the Fe$_3$/Cr$_{14}$/Sn/Cr$_2$ system. One of them is originated from the antiferromagnetic order in the bulk Cr, and the other is connected with incommensurate spin density wave (ISDW) in Cr. A characteristic feature of this system is realization of ISDW in the Cr film thinner than the half length period of a common ISDW in Cr. In the Cr layers with width lower than a quarter of the wave, the ISDW cannot be the cause of the sharp drop of the HFF at Sn. The calculations of the system Fe$_3$/Cr$_8$/Sn/Cr$_8$ with and without allowance for the roughness at the Fe/Cr interface showed that the roughness leads to a significant decrease in the HFF at Sn.

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I. INTRODUCTION

Since the discovery of the antiferromagnetic coupling between Fe layers [1] and giant magnetoresistance [2] in Fe/Cr multilayers a lot of studies have paid a great attention to the Cr magnetic properties in these systems. Up to now, however, the discussion on the magnetic order in thin Cr films continues, there is no clarity in understanding of the interconnection between the Cr magnetic state and the magnetic coupling of Fe layers. Recently, the authors of [3] succeeded in obtaining the Fe/Cr multilayer systems with a Sn monolayer inserted. Multilayer systems Fe$_{d_1}$/Cr$_{d_2}$/Sn/Cr$_{d_3}$ with different widths $d_1$, $d_2$ and $d_3$ were investigated by various physical methods including the Mössbauer spectroscopy. One of the most important results was the established dependence of the hyperfine magnetic field (HFF) at Sn nuclei on the width of the Cr layer. This dependence is characterized by a sharp drop of the HFF magnitude at a Cr width less than 3 nm ($d_2 + d_3 < 3$ nm). Mössbauer experiments cannot give a direct answer as to the magnetization of Cr atoms, but if the HFF at Sn is assumed to be proportional to the local magnetic moments of the nearest Cr atoms, one should inevitably conclude that at a width of the Cr layer less than 3 nm the Cr magnetic moments in these systems are close to zero.

As soon as the experimental data had been available, theoretical first-principles calculations of the electron structure and magnetic characteristics of Fe/Cr/Sn/Cr were conducted [3, 4, 5]. The calculations were performed by both linear methods [3, 4, 5] and the Green function formalism [6] with different approximations of the exchange-correlation potential. As a whole, the results obtained by different methods and authors agree well with each other and do confirm the possibility of estimation of the Cr magnetization from Mössbauer experiments at Sn [6]. Some differences between the results have, though, led to difficulties in explanation of the peculiarities of the HFF variation and elicited a number of unsolved questions.

In the paper presented, we concentrated our attention on the discussion of two hypotheses advanced earlier in [4, 5] for explanation of the Sn HFF behavior. The first one is connected with the existence of the high-spin and low-spin states and a transition between them with a change in the Cr width. Such solutions were obtained for the Fe$_3$/Cr$_{14}$/Sn/Cr$_2$ system in [4]. The second hypothesis is connected with the effect of the Fe/Cr boundary roughness on the Cr magnetization.

We have performed additional calculations of the periodical systems Fe$_3$/Cr$_{14}$/Sn/Cr$_2$ and Fe$_3$/Cr$_8$/Sn/Cr$_8$ (the latter being calculated with and without account of the roughness of the Fe/Cr interface). The calculations were conducted by the following methods: the full-potential linearized augmented plane wave method (FP LAPW) realized in the program package WIEN2k [8], and a screened Korringa-Kohn-Rostoker method developed in Jülich [9] in the atomic sphere approximation (ASA SKKR) and in a full-potential scheme (FP SKKR). The calculations employ the exchange-correlation potential in the local density approximation (LDA) in the formalism of Refs. [10] and [11]. As shown in Ref. [12], if scaled to the moment, the LDA and the generalized gradient approximation (GGA) yield the same behavior.

For our calculations we have used the following models. The multilayer systems were presented as alternating layers in the (001) plane of a bcc lattice with two types of roughness (Fig. 1A and B) and without it. The interlayer distances Fe-Fe, Fe-Cr, Cr-Cr and Cr-Sn were taken equal to 0.144 nm, which corresponds to the half lattice
As stated earlier, the calculation of the Fe$_9$/Cr$_{14}$/Sn/Cr$_2$ system by the SKKR method gave two solutions [6] that we called "high-spin" (HS) and "low-spin" (LS) in accordance with the magnetic moment magnitudes (the difference in energy between the two solutions did not exceed 1 mRy per unit cell). In connection with these solutions it is appropriate to raise here some questions. First: why the LS solution was not found in Ref. [6] by the LAPW method? Second: do there exist two solutions in other systems and do they give a possibility to describe the peculiarities of the Sn HFF behavior as a transition from the HS to the LS state with a decrease of the Cr width? And the third question concerns the nature of the LS state (the character and peculiarities of the behavior of the Cr magnetic moments in the HS solution were considered in detail and explained in [6]).

Two solutions were also found in an isolated 23-layer Cr film by other authors [13]. One of them was classified as a spin density wave and was connected with incommensurate spin density wave (ISDW) of bulk Cr. The ISDW has been studied from sixties, and now reliable experimental and theoretical results are available for both bulk Cr, and some multilayer films with Cr, the majority of which are reflected in reviews [14, 15] in a rather full measure. We did not meet in literature with ISDW in Cr films thinner than 20-22 layers. This seems natural as the ISDW half period exceeds 20 layers. A hypothesis has, nevertheless, arisen that the nature of the low-spin state obtained in [6] is connected with ISDW. For the existence of the ISDW solution, the Fermi-surface topology and the accuracy of its description are important. Assuming that in the FP LAPW calculation of the work [6] the accuracy of the Fermi-surface description was insufficient, we have increased the number of basis wave functions limiting it by the parameter $R_{MT K_{max}} = 7.5$ (in [6] it was $R_{MT K_{max}} = 7.0$). The expansion of the wave functions over the angular momentum was done up to $l_{max} = 10$, and the Fourier expansion of the electron charge density was conducted up to $G_{max} = 20$.

We should note that in the modification of the method used here, namely, the augmented plane wave plus local orbitals (APW+l.o.), the number of plane waves is usually limited by $R_{MT K_{max}} = 7.0$, as the convergence is achieved at lower number of basis functions than in the common realization of the LAPW method [16]. We have increased also the number of k-points in the irreducible part of the Brillouin zone up to 42, which may be crucial for the description of the Fermi surface topology. The calculation was conducted in a scalar-relativistic approximation for the valence electrons and it was fully relativistic for the core electrons with an energy boundary at -7 Ry separating these two groups.

The results of calculation of the Fe$_9$/Cr$_{14}$/Sn/Cr$_2$ system are given in Table I and Fig. 2. The calculations were conducted with a charge convergence criterion $10^{-4} \div 10^{-5}$ (this is a squared charge-density difference between the last two iterations integrated over the space). As seen in Fig. 2 in the FP LAPW method at the chosen parameters we have succeeded in obtaining two solutions, one of them being the LS solution. The HS solution is practically the same as in [6] by the FP LAPW method. The difference in energy between the HS and LS solutions is 1.4 mRy per cell. We should mention a very slow convergence of the LS solution, which prevented us from obtaining the accuracy over spin density of $10^{-5}$ as for the HS solution (the accuracy of $10^{-4}$ for LS was obtained).

The LS solution obtained here has a peculiarity absent in the solution obtained by the SKKR method in Ref. [6], it is a phase slip between the 13-th and 14-th Cr atoms (if counted from the Fe layer, see Table I). The "phase slip" usually denotes a violation of the alternation of the magnetic moment directions characteristic for the antiferromagnetic ordering, and reveals itself in the fact that two neighboring Cr layers have equally directed magnetic
TABLE I: The magnetic moments ($\mu_B$) in Fe$_9$/Cr$_{14}$/Sn/Cr$_2$.

| Atom | layer | HS   | LS   |
|------|-------|------|------|
| Fe   | 2.260 | 2.277|
| Fe   | 2.317 | 2.326|
| Fe   | 2.343 | 2.346|
| Fe   | 2.382 | 2.386|
| Fe   | 2.101 | 2.087|
| Cr   | -0.659| -0.635|
| Cr   | 0.566 | 0.437|
| Sn   | 0.005 | 0.000|
| Cr   | 0.614 | -0.007|
| Cr   | -0.426| -0.014|
| Cr   | 0.530 | 0.050 |
| Cr   | -0.507| -0.106|
| Cr   | 0.545 | 0.157 |
| Cr   | -0.531| -0.229|
| Cr   | 0.560 | 0.284 |
| Cr   | -0.551| -0.343|
| Cr   | 0.565 | 0.386 |
| Cr   | -0.557| -0.438|
| Cr   | 0.566 | 0.463 |
| Cr   | -0.517| -0.458|
| Cr   | 0.500 | 0.443 |
| Cr   | -0.546| -0.522|
| Fe   | 2.034 | 2.021|
| Fe   | 2.429 | 2.429|
| Fe   | 2.330 | 2.343|
| Fe   | 2.340 | 2.352|

From Fig. 3 one may see that SKKR solutions also have a phase slip characteristic for the ISDW (between the 10-th and 11-th sites in ASA SKKR and between the 11-th and 12-th sites in FP SKKR). Note that here we did not succeed in obtaining the second, HS, solution by the SKKR method with an accuracy better than in [8]. Fig. 4 shows the calculation results at two temperatures (400 and 800 K). We should note that to describe the magnetic state variations with temperature (including the phase transitions), the Heisenberg excitations should be allowed for. Our calculations take into account, with the help of the temperature Fermi distribution function, only Stoner-type excitations, which become effective at temperatures as high as 5000–10000 K [17]. This allows us to suppose that, at 400 and 800 K
used, the Stoner excitations weakly affect the magnetic moment magnitude, and to consider the temperature as a mean to change the details of the Fermi surface that determine the position of the phase slip. Indeed, from simple considerations, we may suppose that the ISDW period is determined by the position of the chemical potential which depends on the temperature (see, for example, [18])

\[ \mu = \epsilon_F - \pi^2 k_B T n'(\epsilon_F)/6n(\epsilon_F) + o(k_B T/\epsilon_F) \]

where \( \epsilon_F \) is the Fermi energy, \( k_B \) is the Boltzmann constant, \( n(\epsilon_F) \) and \( n'(\epsilon_F) \) are the electron density of states at the Fermi level and its derivative (the value of \( n'(\epsilon_F) \) is negative in our system). With increasing temperature, the chemical potential grows, decreasing thus the distance in the k-space between the electron and hole surfaces which determines the ISDW period. That is why the phase slip moves from the 10-11-th sites to the 11-12 sites with the temperature change from 400 to 800 K (see Fig. 1).

The above results leave no doubt that the nature of the LS solution in the Fe\textsubscript{8}/Cr\textsubscript{14}/Sn/Cr\textsubscript{2} system is connected with the ISDW state in Cr. The realization of this state in our system has, however, some features absent in the ISDW in other systems [14, 15]. The Sn layer present in the system gives the possibility to form a state with a large magnetic moment connected with the ISDW in other systems [14, 15]. The Sn layer present in our case is the fact that the Sn layer promotes the realization of this state in our system has, however, some features absent in the ISDW in other systems [14, 15]. The Sn layer present in our system gives the possibility to form a state with a large magnetic moment connected with the ISDW in other systems [14, 15]. The Sn layer present in our case is the fact that the Sn layer promotes the realization of this state in our system has, however, some features absent in the ISDW in other systems [14, 15]. The Sn layer present in our case is the fact that the Sn layer promotes the realization of this state in our system has, however, some features absent in the ISDW in other systems [14, 15]. The Sn layer present in our case is the fact that the Sn layer promotes the realization of this state in our system has, however, some features absent in the ISDW in other systems [14, 15]. The Sn layer present in our case is the fact that the Sn layer promotes the realization of this state. This does not explain full measure the experimental sharp drop of the HFF. To obtain it, one should perform the calculation of the systems with higher width and different roughnesses.

### Summary

The first-principles FP LAPW calculation of the multilayer model system Fe\textsubscript{8}/Cr\textsubscript{14}/Sn/Cr\textsubscript{2} gives two solutions. One of them, with a lower magnitude of the Cr magnetic moments and a phase slip, was identified as an ISDW. This identification was confirmed by the SKKR calculations. A characteristic feature of the ISDW state in our case is the fact that the Sn layer promotes the realization of the ISDW state in the films with Cr width less than the half period of ISDW.

In connection with the above statements, one may assert that the band calculations of the periodical multilayer systems without the interface roughness cannot explain the experimentally observed sharp drop of the HFF at Sn with a decrease of the Cr layer width.

The presence of roughness in the Fe/Cr interfaces leads to an HFF drop in thin films. This is confirmed by the

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**TABLE II:** The HFF at Sn (T) and magnetic moments (\( \mu_B \)) in Fe\textsubscript{8}/Cr\textsubscript{8}/Sn/Cr\textsubscript{8}

| Atom | layer | ideal | A roughness (see Fig. 1) | B roughness |
|------|-------|-------|--------------------------|-------------|
| Sn   | 11    | 0.01  | 0.004 / 0.004            | 0.004 / 0.004 |
| Cr   | 1     | 0.73  | 0.23                     | 0.23        |
| Cr   | 2     | -0.48 | -0.15 / -0.15\textsuperscript{a} | -0.15 / -0.15\textsuperscript{a} |
| Cr   | 3     | 0.60  | 0.18                     | 0.18        |
| Cr   | 4     | -0.57 | -0.18 / -0.18\textsuperscript{a} | -0.16 / -0.18\textsuperscript{a} |
| Cr   | 5     | 0.60  | 0.19                     | 0.17        |
| Cr   | 6     | -0.55 | -0.18 / -0.24\textsuperscript{a} | -0.15 / -0.22\textsuperscript{a} |
| Cr   | 7     | 0.54  | 0.23                     | 0.19        |
| Cr / Fe | 8  | -0.60 | -0.53 / 1.92\textsuperscript{a} | -0.50 / 1.89\textsuperscript{a} |
| Fe   | 9     | 1.97  | 2.25                     | 2.25        |
| Fe   | 10    | 2.46  | 2.43                     | 2.37 / 2.47\textsuperscript{a} |

\( \text{.../... denotes values for two nonequivalent atoms in the layer} \)
first-principles FP LAPW calculations conducted for the model $Fe_3/Cr_8/Sn/Cr_8$ with ideal Fe/Cr interfaces and with two types of roughness. The calculations showed that there is a threefold drop in HFF in the systems with roughness as compared to the system with ideal Fe/Cr boundaries. However for explanation of the experimental dependence it is necessary to calculate a number of systems with more than 20-23 Cr layers and with allowance for the different roughness.

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