PHYSICAL REVIEW B VOLUME 59, NUMBER 10 1 MARCH 1999-II

Spin-density wave in Fe/Cr superlattices: A first-principles study

Kunitomo Hirai
Department of Physics, Nara Medical University, Kashihara, Nara 634-8521, Japan
(Received 17 November 1998)

A first-principles electronic-structure calculation for Fe/Cr superlattices is presented, where a spin-density-wave order in the Cr layer is considered in addition to an antiferromagnetic one. The interlayer magnetic coupling between ferromagnetic Fe layers is investigated, and the oscillation of the interlayer magnetic coupling with a two-monolayer period of the spacer thickness of the Cr layer is illustrated. The appearance of the spin-density-wave order in the Cr layer, which gives rise to a phase slip of the oscillation, is furthermore demonstrated. [S0163-1829(99)51010-2]

As an archetype of spin-density wave (SDW) in itinerant electron systems, Cr and its alloys have been extensively investigated up to the present.[1] An interest has recently focused on SDW in a Cr spacer of Fe/Cr superlattices, in connection with the interlayer magnetic coupling between ferromagnetic Fe layers.[2] The coupling of magnetizations of two successive Fe layers oscillates between parallel and antiparallel with a two-monolayer period of the spacer thickness of a Cr layer, and for thicker Cr layers the oscillation is followed by periodic phase slips, as is revealed by scanning electron microscopy with polarization analysis (SEMPA) studies.[3] These phase slips are supposed to be due to incommensurability of an SDW order in the Cr layer, and the appearance of an SDW order is ascertained by neutron-diffraction studies for the superlattices with a relatively thick Cr layer of more than 36 monolayers.[4] The SDW order in Fe/Cr superlattices is thus substantial when the spacer thickness of a Cr layer is large enough. On the other hand, it is generally accepted, at least in a theoretical aspect, that the Cr layer has a nonmagnetic or antiferromagnetic (AF) order when the spacer thickness of a Cr layer is small;[5] either of the orders roughly accounts for the oscillation of the interlayer magnetic coupling.[6] The nonmagnetic order, which may be justified by the existence of interfacial roughness,[7] has a magnetism induced by the proximity Fe layer.[8] The AF order, to the contrary, is inherent in the Cr layer itself and is expected to appear when the spacer thickness is small compared with the period of the SDW, as is suggested in electronic structure calculations for Fe/Cr superlattices.[7] The question to be discussed here is what the magnetic order in the Cr layer is, specifically, whether the order is AF or of SDW, when the spacer thickness of a Cr layer is intermediate.[9]

There are two points at issue; one is a critical thickness of a Cr layer above which the SDW order appears, and the other is whether the SDW in Fe/Cr superlattices is attributed to the proximity Fe layer or to the Fermi-surface nesting responsible mostly to the Cr layer, which is intrinsic in the bulk Cr. These points have so far been discussed qualitatively by means of a phenomenological model,[10] but they have hardly been discussed quantitatively, although the Fermi-surface nesting, which is closely related to its electronic structure, is a seriously delicate factor. The electronic structure calculation of first principles, which is at present most reliable, is therefore expected to supply significant knowledge for the quantitative discussion on the SDW in Fe/Cr superlattices. In most of the electronic structure calculations for Fe/Cr superlattices, however, the AF order has been considered as the magnetic order in the Cr layer. The SDW order has never been considered properly, since the calculation for the SDW order is rather time consuming on account of the large number of atoms in the unit cell;[11] a calculation for the SDW order was once done without fulfillment of a self-consistent procedure,[12] but it is questionable whether such a non-self-consistent procedure is competent to discuss a delicate stability between the AF and SDW orders.

In this paper, we present a first-principles electronic structure calculation for Fe/Cr superlattices, which is performed by means of the Korringa-Kohn-Rostoker (KKR) Green's-function method within the framework of the local spin density (LSD) functional formalism. The calculation is a self-consistent one without adjustable parameters and is carried out for periodic superlattices which consist of ferromagnetic Fe layers and AF or SDW Cr layers, with the magnetizations of two successive Fe layers being aligned parallel or antiparallel. The purpose of this work is to supply reliable knowledge for the quantitative discussion on the SDW in Fe/Cr superlattices. We survey variation of the interlayer magnetic coupling with respect to the spacer thickness of a Cr layer, together with an investigation for the possibility of the appearance of an SDW order in the Cr layer. We add that the present author already performed a first-principles calculation for bulk SDW Cr with results in good agreement with experiments concerning its magnetism; for example, the wave vector of SDW of the lowest total energy per atom is found to be $a^*(19/20,0,0)$, which is close to the observed one at low temperature $a^*(0.952,0,0)$, where $a^*=2\pi/a$ and a denotes a lattice constant of the chemical bcc lattice.[13]

Through self-consistent calculations with different initial magnetic orders in the Cr layer, we can confirm that the coupling of the local magnetic moments between Fe and Cr atoms across the interface is a strongly antiparallel one in comparison with that between Cr atoms, which may be readily understood in the case of the superlattices without interfacial roughness.[7] The strong interfacial coupling between Fe and Cr layers influences the magnetic order in the inner Cr layer, especially when Fe layers exist on both sides of the Cr layer, as is the case of periodic Fe/Cr superlattices

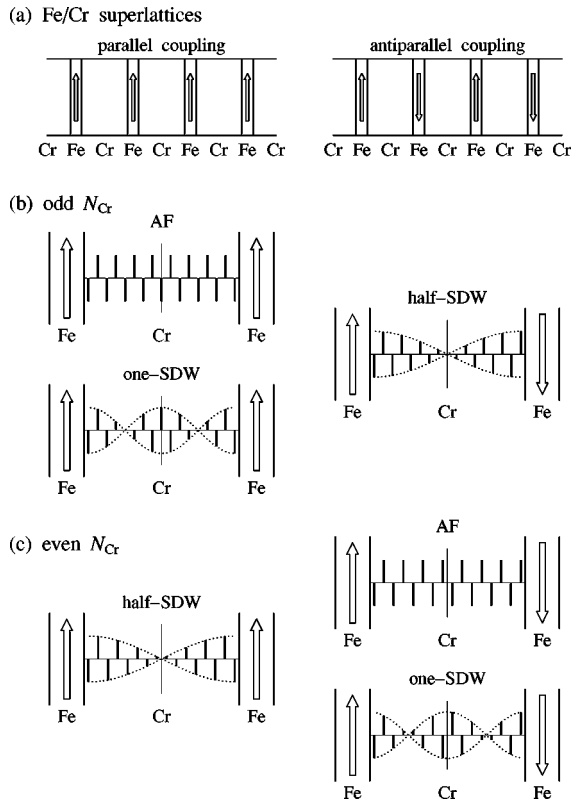

FIG. 1. Magnetic order in (a) Fe/Cr superlattices and in the spacer-Cr layer for the cases of (b) odd N_{Cr} and (c) even N_{Cr}. An arrow indicates the direction of the magnetization of an Fe layer, and a bar indicates the local magnetic moment of a Cr atom. The local magnetic moment of a Cr atom at the interface is antiparallel to the magnetization of the neighboring Fe layer.

or Fe/Cr/Fe sandwiches. This can be seen in Fig. 1 of the magnetic order, and N_{Cr} proves to play a key role, where N_{Cr} denotes the number of monolayers of the spacer Cr layer. When the magnetizations of two successive Fe layers are aligned parallel, an AF order is commensurate within the spacer-Cr layer of an odd N_{Cr}. For the spacer-Cr layer of an even N_{Cr}, however, an AF order needs a defect in a magnetic sense, and we can hardly obtain a self-consistent solution of the calculation if such a defect exists. Instead of an AF order with a defect, we can obtain a self-consistent solution of an SDW order such that the spacer-Cr layer contains a half period of SDW with its antinodes located at interfaces (half-SDW order); the strong interfacial coupling favors a larger magnitude of the local magnetic moment of a Cr atom at the interface. This half-SDW order is really commensurate within the spacer-Cr layer of an even N_{Cr}. To the contrary, when the magnetizations of two successive Fe layers are aligned antiparallel, an AF order is commensurate within the spacer-Cr layer of an even N_{Cr} and a half-SDW order is commensurate within that of an odd N_{Cr}. The difference in the half-SDW order between two cases of odd and even N_{Cr} consists in the presence of a nodal monolayer; there is really a nodal monolayer at the center of the Cr layer for an odd N_{Cr} while not for an even N_{Cr}.

The magnetic order in the Cr layer is thus governed by whether N_{Cr} is even or odd, that is, the parity of N_{Cr}, and the oscillation of the interlayer magnetic coupling with a two-monolayer period seems quite natural. There is however a difference between the AF and half-SDW orders in the correspondence of the parity of N_{Cr} with the coupling of the magnetizations of the Fe layers. In addition to the half-SDW order, we have an SDW order such that the spacer-Cr layer contains one period of SDW (one-SDW order), and this one-SDW order is commensurate within the spacer-Cr layer under the same situation as the AF order (see Fig. 1). Because of the strong interfacial coupling, the antinode of SDW is generally fixed on the interface, and SDW orders commensurate within the spacer-Cr layer may be restricted to half-SDW, one-SDW, $\frac{3}{2}$-SDW, and so on, when the spacer thickness of a Cr layer, that is, N_{Cr} is not so large.[14] It is to be emphasized that such commensurability within the spacer Cr layer is not an assumption but a consequence of the self-consistent calculations. The wave vector of the SDW orders is thus not determined by the Fermi-surface nesting but by N_{Cr}, so long as N_{Cr} is not so large; for example, the half-SDW order corresponds to an SDW order of a wave vector $a^*(q,0,0)$ with $q=(N_{Cr}-2)/(N_{Cr}-1)$ and accordingly to that of bulk Cr when N_{Cr} is $20\sim24$. When N_{Cr} is not so large, it is not likely that the Cr layer in superlattices admits an incomplete portion of SDW with a specific wave vector, which is usually determined by the Fermi-surface nesting and is in principle incommensurate with the underlying lattice.

Let us discuss results of the calculation, which is carried out for the superlattices with $N_{Cr}\leqslant 21$ and with $N_{Fe}=3$ for an odd N_{Cr} and $N_{Fe}=4$ for an even N_{Cr}, where N_{Fe} is the number of monolayers of an Fe layer. We adopt an experimental lattice constant of bulk Cr, that is, $a=5.45$ a.u. to view variation of the interlayer magnetic coupling between Fe layers J_{Fe} with respect to N_{Cr}; we assume that the superlattice retains cubic lattice spacing. Here J_{Fe} is defined as

$$J_{Fe}=E_{ap}-E_{p}, \tag{1}$$

where E_{ap} and E_{p} are the total energy per one atom when the magnetizations of the two successive Fe layers are aligned antiparallel and parallel, respectively, and the AF and half-SDW orders in the Cr layer are considered.[15] Figure 2 shows J_{Fe}, and it is found that J_{Fe} is positive (parallel coupling is favored) for an odd N_{Cr} and negative (antiparallel coupling is favored) for an even N_{Cr}. The result is consistent with the fact that the interlayer magnetic coupling oscillates with a two-monolayer period, and it means that in the Cr layer the AF order is more favorable than the half-SDW one, in so far as $N_{Cr}\leqslant 21$, though the energy difference between the AF and half-SDW orders becomes smaller as N_{Cr} becomes large. The calculation for the case of $a=5.45$ a.u. thus illustrates the oscillation of J_{Fe} with a two-monolayer period but does not indicate the appearance of the SDW order in the Cr layer.

The conclusion mentioned above, however, is drawn from the result of the calculation for the case of the experimental lattice constant. For a crucial discussion concerning the stable magnetic order, we have to discuss the case of an equilibrium lattice constant a_0 at which the total energy becomes minimum. We therefore carry out the calculation with varying lattice constant a to determine a_0, for two cases of $N_{Cr}=9$ and $N_{Cr}=19$. In Fig. 3, we show E_p, E_{ap}, J_{Fe}, and the magnitude of the local magnetic moment of a Cr atom at the interface m_{Cr}^{if}, as a function of a. It is found that E_p and

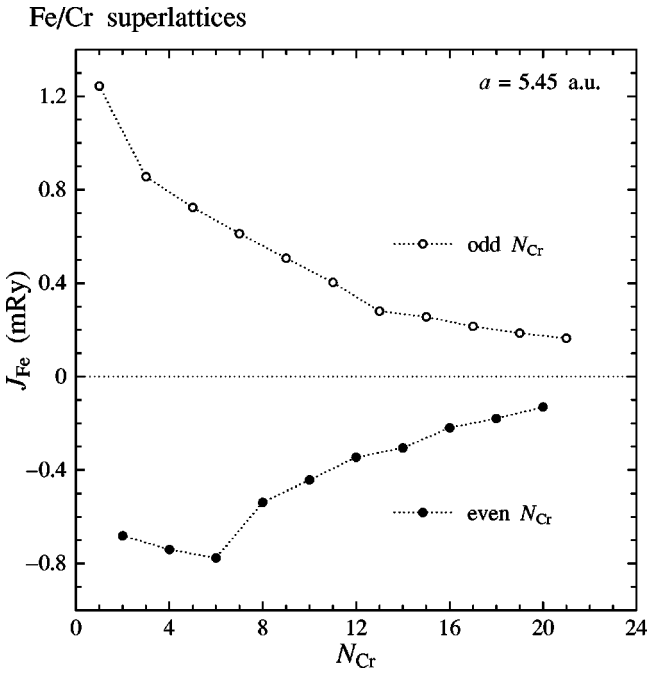

FIG. 2. Variation of the interlayer magnetic coupling between Fe layers J_{Fe} with respect to the spacer thickness of the Cr layer. Here J_{Fe} for the case of $a=5.45$ a.u. is shown as a function of N_{Cr}.

E_{ap} both become minimum at around $a=5.32$ a.u., that is, $a_0 \simeq 5.32$ a.u. for both cases of $N_{Cr}=9$ and $N_{Cr}=19$; this value is about 98% of the experimental value, which is common with the use of the LSD formalism.[16] The curves of E_p and E_{ap} do not cross as a varies for the case of $N_{Cr}=9$, but they cross each other for the case of $N_{Cr}=19$, with a reversal of the sign of J_{Fe}. At the equilibrium lattice constant a_0, J_{Fe} is positive for $N_{Cr}=9$ while negative for $N_{Cr}=19$, which means that in the Cr layer the AF order is favorable for $N_{Cr}=9$ whereas the half-SDW order is favorable for $N_{Cr}=19$. This is different from the case of $a=5.45$ a.u., where the AF order is always favorable. The difference between these two cases of a can be ascribed to the magnitude of the local magnetic moment of a Cr atom. As can be seen in Fig. 3(c), for the case of $a=5.45$ a.u., we have a larger magnitude of the local magnetic moment of a Cr atom in comparison with that for the case of $a=a_0$, which usually makes the AF order favorable.[17] When the magnitude of the local magnetic moment of a Cr atom is not so large and N_{Cr} is around 20, where the half-SDW order approaches the SDW order of bulk Cr, an energy gain due to the nesting mechanism may become dominant to make the half-SDW order favorable. Thus the calculation for the case of the equilibrium lattice constant surely indicates the appearance of the SDW order in the Cr layer.

Although the calculation to determine the equilibrium lattice constant a_0 is only performed for $N_{Cr}=9$ and 19, we can sufficiently discuss the variation of J_{Fe} with respect to N_{Cr} for the case of $a=a_0$, with particular attention to the phase slips of the oscillation of the interlayer magnetic coupling observed in SEMPA. We expect that the oscillation of J_{Fe} with a two-monolayer period basically does not change, since the two-monolayer period is originated from the parity of N_{Cr}, in other words, in consequence of the commensurability within the spacer-Cr layer of the AF and SDW orders.

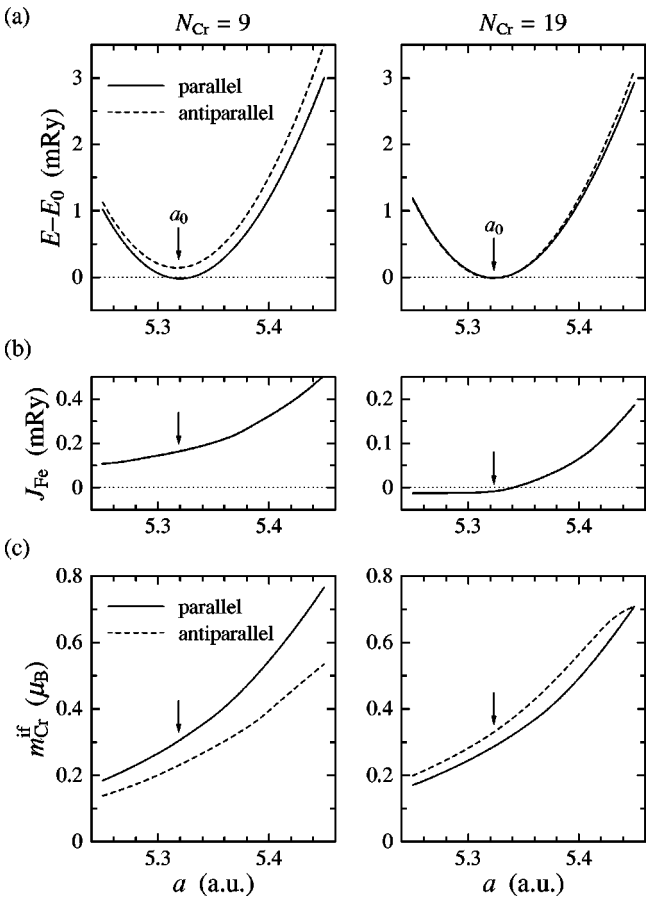

FIG. 3. Dependence of the total energy and the magnitude of the local magnetic moment of a Cr atom upon the lattice constant a for the cases of $N_{Cr}=9$ and 19. Here (a) E_p and E_{ap}, (b) their difference J_{Fe}, and (c) m_{Cr}^{if} are shown, where the reference energy E_0 is E_p of $a=5.33$ a.u. A vertical arrow indicates the position of the equilibrium lattice constant a_0.

We may furthermore expect that there is a critical N_{Cr}, which is denoted by N_{Cr}^c, between 9 and 19; below N_{Cr}^c, the AF order is more favorable than the half-SDW one, and above N_{Cr}^c, the half-SDW order is more favorable than the AF one. At N_{Cr}^c, the relative stability between the AF and half-SDW orders is reversed, and accordingly correspondence between the parity of N_{Cr} and the sign of J_{Fe} is also reversed; that is, when $N_{Cr}<N_{Cr}^c$, $J_{Fe}>0$ for odd N_{Cr} and $J_{Fe}<0$ for even N_{Cr}, whereas when $N_{Cr}>N_{Cr}^c$, $J_{Fe}<0$ for odd N_{Cr} and $J_{Fe}>0$ for even N_{Cr}. This reversal of the correspondence gives rise to a phase change by π in the oscillation of J_{Fe}, which is nothing but a phase slip of the oscillation. Such a phase slip is also expected to appear at another critical N_{Cr} for the stability between the half-SDW and one-SDW orders or such, since the stable magnetic order in the Cr layer will change as AF, half-SDW, one-SDW, $\frac{3}{2}$-SDW, . . . , when N_{Cr} increases. The phase slips are thus irrelevant to incommensurability of the SDW order itself, as is shown in the discussion based on a phenomenological model.[10] Here we will not discuss values of N_{Cr} of the phase slips, the reported values of which are 24, 44, and 64,[3] since the present calculation has not been completed yet; the discussion will be presented in a future publication, with respect to effects of temperature or interfacial roughness.

Finally, we briefly mention a helical (or spiral) SDW order in the Cr layer, which is a noncollinear magnetic order, unlike the sinusoidal one that we have so far considered. In bulk Cr, the helical SDW order never appears, since the energy gain due to the nesting mechanism of the helical SDW order is always smaller than that of the sinusoidal one with the same wave vector.[1,17] Similarly, in the Fe/Cr superlattices, it is not likely that the helical SDW order becomes more stable than the sinusoidal one when the magnetizations of the Fe layers are aligned parallel or antiparallel, that is, collinear. However, when the magnetizations of the Fe layers are not collinear,[18] there seems a possibility of the appearance of the helical SDW order, in which the local magnetic moments of the Fe and Cr atoms across the interface can be aligned antiparallel; the helical SDW order possibly does not suffer so large an energy loss caused by compulsory twisting of the local magnetic moments as the sinusoidal one suffers. We hope that the helical SDW order might be stabilized in the Fe/Cr superlattices under an optimum condition for the angle between the magnetizations of the Fe layers and the spacer thickness of the Cr layer;[19] for example, in the case where the magnetizations of two successive Fe layers are perpendicular, a helical SDW order with a quarter (or $\frac{3}{4}, \frac{5}{4}, \ldots$) period will have an energy gain due to the nesting mechanism and might be stabilized when N_{Cr} is around 11 (or 33,55,...).

In conclusion, we present a first-principles electronic-structure calculation for the SDW in Fe/Cr superlattices for the first time. We investigate the interlayer magnetic coupling between ferromagnetic Fe layers in the Fe/Cr superlattices with respect to the spacer thickness of the Cr layer. It is shown that the interlayer magnetic coupling oscillates with a two-monolayer period of the spacer thickness, which may be due to the commensurability within the spacer-Cr layer of the AF and SDW orders. It is also demonstrated that an SDW order can appear in the Cr layer when N_{Cr} is large (at least, when $N_{\mathrm{Cr}}=19$), and the phase slip in the oscillation of the interlayer magnetic coupling is discussed in connection with the appearance of the SDW order.

The author would like to express his sincere thanks to Professor H. Akai for helpful discussion concerning the KKR-LSD calculation. The author thanks the Supercomputer Center, Institute for Solid State Physics, the University of Tokyo, for the use of the FACOM VPP500.

[1] E. Fawcett, Rev. Mod. Phys. **60**, 209 (1988); E. Fawcett, H. L. Alberts, V. Yu. Galkin, D. R. Noakes, and J. V. Yakhmi, *ibid.* **66**, 25 (1994).

[2] A. Fert, P. Grünberg, A. Barthélémy, F. Petroff, and W. Zinn, J. Magn. Magn. Mater. **140-144**, 1 (1995).

[3] J. Unguris, R. J. Celotta, and D. T. Pierce, Phys. Rev. Lett. **69**, 1125 (1992).

[4] E. E. Fullerton, S. D. Bader, and J. L. Robertson, Phys. Rev. Lett. **77**, 1382 (1996).

[5] The nonmagnetic order in the Cr layer is still controversial, though it is addressed in some experiments, for example, J. Meersschaut, J. Dekoster, R. Schad, P. Beliën, and M. Rots, Phys. Rev. Lett. **75**, 1638 (1995).

[6] S. Mirbt, A. M. N. Niklasson, B. Johansson, and H. L. Skriver, Phys. Rev. B **54**, 6382 (1996).

[7] D. Stoeffler and F. Gautier, J. Magn. Magn. Mater. **121**, 259 (1993); **147**, 260 (1995).

[8] J. C. Slonczewski, Phys. Rev. Lett. **67**, 3172 (1991).

[9] Terminology of AF and SDW is sometimes confused. The present way of discrimination of these two terms is the following; there is no node in the AF order, while there is at least one node in the SDW order.

[10] Z. P. Shi and R. S. Fishman, Phys. Rev. Lett. **78**, 1351 (1997); R. S. Fishman, and Z. P. Shi, J. Phys.: Condens. Matter **10**, L277 (1998).

[11] A real-space method of electronic structure calculation adopted in Ref. 7 is not an appropriate one for the purpose of discussing the SDW order, since the method cannot appreciate the Fermi-surface nesting.

[12] M. van Schilfgaarde and F. Herman, Phys. Rev. Lett. **71**, 1923 (1993).

[13] K. Hirai, J. Phys. Soc. Jpn. **66**, 560 (1997).

[14] When N_{Cr} becomes large, the antinode of these SDW orders may sometimes move away from the interfacer, with the period of the SDW orders deviating from a multiple of a half.

[15] The one-SDW order will be included in the calculation if we consider a case of $N_{\mathrm{Cr}}>21$.

[16] K. Hirai, J. Phys. Soc. Jpn. **67**, 1776 (1998).

[17] K. Hirai, J. Phys. Soc. Jpn. **62**, 690 (1993); **65**, 586 (1996).

[18] The noncollinear magnetic coupling in Fe/Cr superlattices is actively investigated now, for example, A. Schreyer *et al.*, Phys. Rev. Lett. **79**, 4914 (1997).

[19] This is one mechanism of the noncollinear magnetic coupling peculiar to Cr superlattices, which has not been discussed in J. C. Slonczewski, J. Magn. Magn. Mater. **150**, 13 (1995).