Spontaneous spin-density-wave order in Cr superlattices

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Spin-density-wave order in Cr superlattices with a relatively thick Cr layer is discussed on the basis of a first-principles electronic structure calculation for those with a boundary layer of ferromagnetic Fe or nonmagnetic V. The spin-denisty-wave order undergoes large influence of proximity effects of the boundary layer, around the interface between the Cr and boundary layers. When a Cr layer is sufficiently thick, however, the spin-density-wave order grows spontaneously and becomes similar to that in bulk Cr, in the vicinity of the middle of the Cr layer.

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Spin-density wave (SDW), which is one of the key concepts of the itinerant electron magnetism, is conspicuous in the respect that its origin is ascribed to a special feature of the Fermi surface, that is, the nesting between electron and hole Fermi surfaces.[1] The SDW order of a long-range modulation in a real space reflects the feature of the electronic structure viewed in a reciprocal space, which is not common to a ferromagnetic or simple antiferromagnetic (AF) order at all.

As the archetype of SDW, Cr has been extensively investigated up to the present,[2] and recently interest has been focused on SDW in Cr superlattices with layered structures, where characteristics of SDW are affected through interfacial magnetic coupling with a ferromagnetic or nonmagnetic boundary layer.[3] The SDW in Cr superlattices may be altered from that in bulk Cr under real-space constraints due to proximity effects of the boundary layer.

Above all, SDW in Fe/Cr/Fe trilayers or Fe/Cr multilayers has attracted much attention,[4] in connection with oscillatory properties of interlayer magnetic coupling between ferromagnetic Fe layers.[5] The interlayer magnetic coupling oscillates between parallel and antiparallel with a two-monolayer period of the spacer thickness of a Cr layer, but the oscillation is followed by periodic phase slips for a thicker Cr layer.[6] A first-principles electronic structure calculation for Fe/Cr superlattices has shown that the two-monolayer period of the oscillation comes from decisiveness of the parity of N_{Cr} (that is, whether N_{Cr} is odd or even), which is due to commensurability of magnetic orders within the Cr spacer layer, where N_{Cr} is the number of monolayers of the Cr layer.[7] Moreover, the calculation has shown that the magnetic order in the Cr layer changes from an AF one to an SDW one as N_{Cr} increases and this change gives rise to a phase slip of the oscillation.

In the correct description, this SDW order in the Fe/Cr superlattices is an order such that the Cr layer contains a half period of SDW (half SDW order) with its antinodes located around the interfaces between the Fe and Cr layers and its node located at the middle of the Cr layer. This is due to strong (antiferromagnetic) interfacial coupling with the Fe magnetization, which favors a larger magnitude of the local magnetic moment of a Cr atom around the interface. The SDW order in the Fe/Cr superlattices, at least the half SDW one, undergoes large influence of proximity effects of the Fe layer and exhibits characteristics different from those of the SDW in bulk Cr. For instance, the wave vector of SDW is not determined by the nesting vector but by the thickness N_{Cr}, and the amplitude of SDW does not disappear even for a very small N_{Cr}, corresponding to a wave vector far away from the nesting one.[8] The magnitude of the local magnetic moment of a Cr atom at the end of the Cr layer $|m_{\mathrm{Cr}}^{\mathrm{end}}|$ becomes almost constant as N_{Cr} increases, which also supports the large influence of the Fe layer.

These facts about the SDW order in the Fe/Cr superlattices motivate us to advance further calculations for Cr superlattices; one is a calculation for the Fe/Cr superlattices with a larger N_{Cr}, where the Cr layer will contain one period of SDW (one SDW order) or one and a half period of SDW (3/2 SDW order), and another is a calculation for superlattices with a nonmagnetic metal like V. The calculations are expected to reveal characteristics of the SDW order in Cr superlattices more clearly and illustrate sensitiveness of the SDW order to its surroundings and N_{Cr}. One of the keen interests is how the SDW order changes from an order governed by proximity layers to an order inherent in the Cr layer itself, which will grow rather spontaneously with a wave vector independent of N_{Cr} and probably resemble the SDW in bulk Cr.

The aim of the present study is to investigate crossover of the SDW order from that due to the proximity magnetism to that intrinsic to the Cr layer. The investigation is fulfilled by a first-principles electronic structure calculation for Fe/Cr and V/Cr superlattices with a sufficiently large N_{Cr}, and we expect that the investigation may provide convincing evidence for the discussion about the SDW order in Cr superlattices.

This study of a first-principles calculation is performed by means of the Korringa-Kohn-Rostoker (KKR) Green-function method within the framework of the local spin-density (LSD) functional formalism. The calculation is carried out for periodic superlattices of bcc(100) which consist of Fe or V layers and Cr layers, where magnetizations of two successive Fe layers or induced ones of V layers are usually assumed to align parallel.[9] A case where the magnetizations align antiparallel is not discussed here, but this may not cause any serious effects for the discussion about the SDW order, as has been proved in the previous calculation (see below).

Let us begin with a brief review of the previous calculation for the Fe/Cr superlattices, in which $N_{\rm Cr}$ is varied up to 21. We have self-consistent solutions of the AF and half SDW orders for odd and even $N_{\rm Cr}$'s, respectively, for the case of parallel magnetizations, while those of the AF and half SDW orders for even and odd $N_{\rm Cr}$'s, respectively, for the case of antiparallel magnetizations. These half SDW orders slightly differ between the two cases of parallel and antiparallel magnetizations, that is, of even and odd $N_{\rm Cr}$'s, but the difference is small and variation of the half SDW order with respect to $N_{\rm Cr}$ is smooth. A fundamental wave of the half SDW order is specified by a wave vector $2\pi/a(q,0,0)$ with $q=(N_{\rm Cr}-2)/(N_{\rm Cr}-1)$, where a is the lattice constant of the bcc lattice, and amplitudes of harmonics of the half SDW order turn out to be fairly small in comparison with an amplitude of the fundamental wave, so long as $N_{\rm Cr}\leqslant 21$.

The present calculation with a much larger even $N_{\rm Cr}$ for the Fe/Cr superlattices shows that the half SDW order undergoes some modulation, that is, its profile becomes more "rectangular" (not "triangular"). This can be noticed in Fig. 1(a), where distribution of the local magnetic moments in the Cr layer, that is, the magnitude of the local magnetic moment of a Cr atom in each monolayer $m_{\rm Cr}^{\nu}$, with $\nu=\nu' -(N_{\rm Cr}+1)/2$ and $\nu'=1,2,\ldots,N_{\rm Cr}$, is shown for $N_{\rm Cr}=36$. In Fig. 1(a), we also show variation with respect to $N_{\rm Cr}$ of $|m_{\rm Cr}^{\rm end}|$, $|m_{\rm Cr}^{\rm mid}|$, or $|m_{\rm Cr}^{\rm quart}|$, where $m_{\rm Cr}^{\rm mid}$ and $m_{\rm Cr}^{\rm quart}$ are $m_{\rm Cr}^{\nu}$ near to the middle and that near to the quarter position of the Cr layer, respectively. It is found that these magnitudes become nearly constant for a larger $N_{\rm Cr}$ and also that $|m_{\rm Cr}^{\rm quart}|$ is larger than $|m_{\rm Cr}^{\rm end}|/\sqrt{2}$ for $N_{\rm Cr}>24$, which indicates rectangular profile of the half SDW order. The modulation of the half-SDW order can be expressed by a Fourier series

$$m_{\rm Cr}^{\nu}=\sum_{n} M_n \cos(\pi q_n \nu), \tag{1}$$

which has nonzero components M_n for wave numbers

$$q_2=\frac{N_{\rm Cr}-2}{N_{\rm Cr}-1},\quad q_4=\frac{N_{\rm Cr}-4}{N_{\rm Cr}-1},\quad q_6=\frac{N_{\rm Cr}-6}{N_{\rm Cr}-1},\ldots, \tag{2}$$

where M_2 corresponds to the amplitude of the fundamental wave of the half SDW order and $M_4, M_6, \cdots$ correspond to the amplitudes of the harmonics. The variation of M_2, M_4, or M_6 is also shown in Fig. 1(a), and it is found that M_4 becomes positive and then increases when $N_{\rm Cr}$ exceeds 20, which is consistent with the appearance of the rectangular profile.

For an odd $N_{\rm Cr}$, the calculation shows that a self-consistent solution of the one SDW order appears for $N_{\rm Cr}\geqslant 33$. This one SDW order has an antinode at the middle of the Cr layer, as can be seen in Fig. 1(b), where the distribution for $N_{\rm Cr}=37$ is shown, together with the variation of $|m_{\rm Cr}^{\rm end}|$, $|m_{\rm Cr}^{\rm mid}|$, and $|m_{\rm Cr}^{\rm quart}|$. It is found that $|m_{\rm Cr}^{\rm mid}|$ is smaller than $|m_{\rm Cr}^{\rm end}|$ and hence the one SDW order is somewhat modulated from that by naive definition of one cosine function.[10] This modulation of the one SDW order can be described as considerable mixing of the AF order, since the

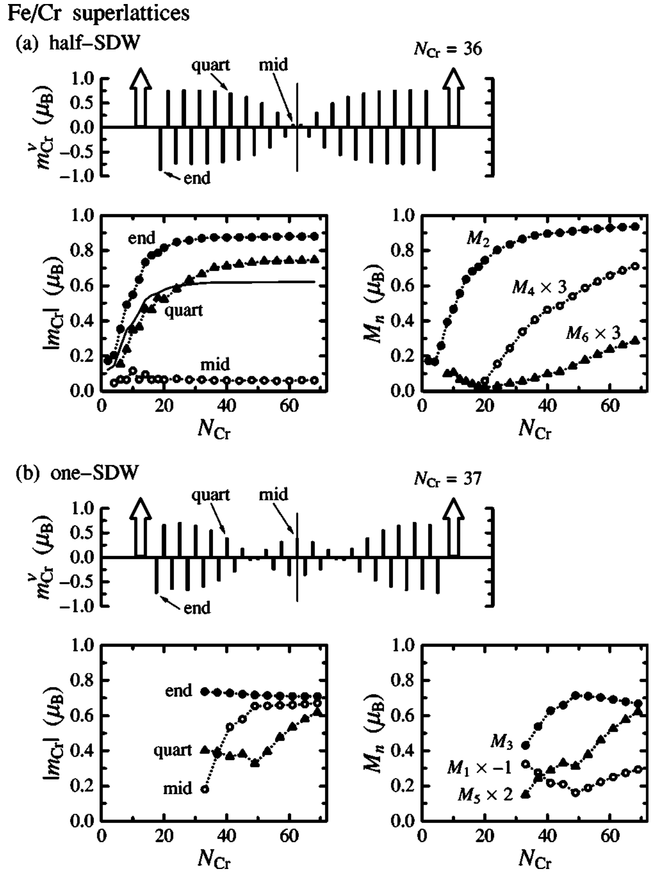

FIG. 1. Distribution of the local magnetic moments in the Cr layer for the (a) half SDW with an even $N_{\rm Cr}$ and (b) one SDW orders with an odd $N_{\rm Cr}$ in the Fe/Cr superlattices, where $m_{\rm Cr}^{\nu}$ is shown with an abscissa of ν. The Fe magnetizations (big arrow) align parallel, and the distribution is symmetric with respect to the middle of the Cr layer (thin vertical line). Variation with respect to $N_{\rm Cr}$ of $|m_{\rm Cr}^{\rm end}|$, $|m_{\rm Cr}^{\rm mid}|$, or $|m_{\rm Cr}^{\rm quart}|$ and that of significant M_n's are shown below the figure of distribution. A solid line neighboring $|m_{\rm Cr}^{\rm quart}|$ for the half SDW order indicates $|m_{\rm Cr}^{\rm end}|/\sqrt{2}$.

Fourier series of Eq. (1) for the one SDW order has significant components M_n for wave numbers

$$q_1=1,\quad q_3=\frac{N_{\rm Cr}-3}{N_{\rm Cr}-1},\quad \text{and}\quad q_5=\frac{N_{\rm Cr}-5}{N_{\rm Cr}-1}, \tag{3}$$

where M_1 corresponds to the amplitude of the AF order and M_3 corresponds to that of the fundamental wave of the one SDW order. From the variation of M_1, M_3, or M_5 shown in Fig. 1(b), it is found that M_1 does not monotonously decrease but increases for $N_{\rm Cr}>49$, where M_3 slightly decreases in contrast.

The variation of the Fourier components M_1 and M_3, and also that of $|m_{\rm Cr}^{\rm quart}|$, may suggest change of the one SDW order for a larger $N_{\rm Cr}$, which can be clearly seen in Fig. 2, where the distribution of the Cr local magnetic moments is shown with $N_{\rm Cr}$ varying. As $N_{\rm Cr}$ increases, an interval from the middle to the node λ_1, four times of which corresponds to the period of SDW, becomes a constant of 10 monolayers (see the lower right of Fig. 2), and similarly, $|m_{\rm Cr}^{\rm mid}|$, which

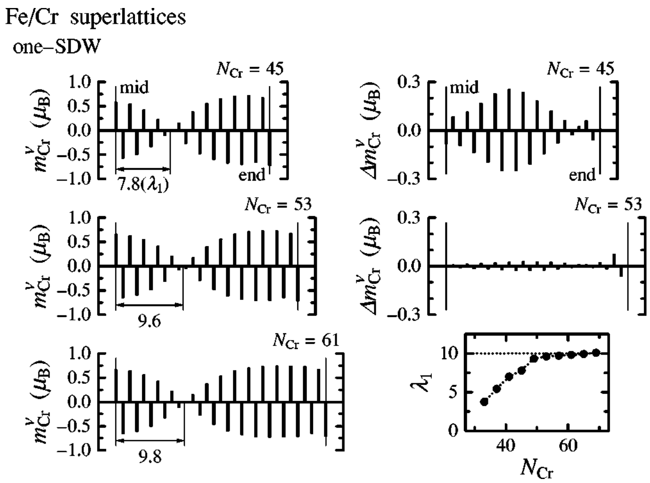

FIG. 2. Change of the one SDW order in the Fe/Cr superlattices, where a right half of the distribution of the Cr local magnetic moments m^{ν}_{Cr} is shown for $N_{Cr}=45,53,61$, and a value under an arrow indicates λ_1. Difference of the distributions Δm^{ν}_{Cr} from that of $N_{Cr}=61$ is also shown, together with variation of λ_1 with respect to N_{Cr}.

corresponds to the amplitude of SDW, becomes a constant of 0.67 μ_B [see Fig. 1(b)]. These values of $10\times4=40$ and 0.67 are both close to those obtained in the calculation for bulk Cr and also to those observed in experiments.[2,8] The distributions for larger N_{Cr}'s become alike to each other, particularly in the vicinity of the middle of the Cr layer, which can be readily confirmed by difference of the distributions Δm^{ν}_{Cr} from that of $N_{Cr}=61$ shown in Fig. 2; the difference between $N_{Cr}=53$ and 61 is fairly small in comparison with that between $N_{Cr}=45$ and 61. The results mentioned so far indicate that the change of the one SDW order takes place at around 49.

Such a change of the SDW order can also be seen in Fig. 3, where figures similar to Fig. 2 are shown for the 3/2 SDW order; a solution of the 3/2 SDW order appears for an even $N_{Cr}\geqslant 44$. As N_{Cr} increases, an interval from the middle to the

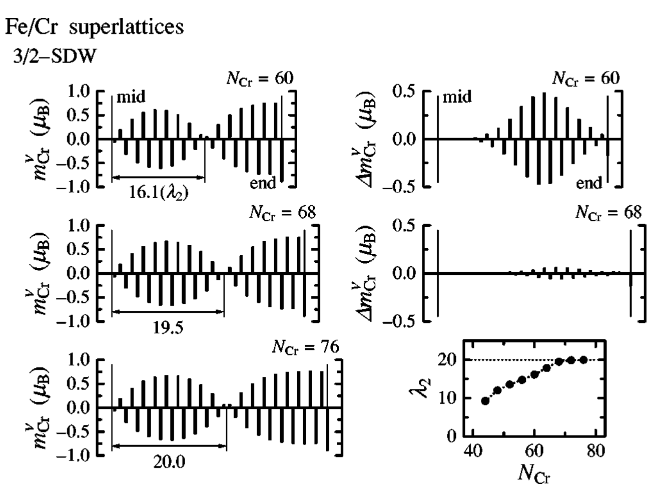

FIG. 3. Change of the 3/2 SDW order in the Fe/Cr superlattices. Others are the same as those in Fig. 2, except that m^{ν}_{Cr} is shown for $N_{Cr}=60,68,76$ and λ_1 is replaced by λ_2.

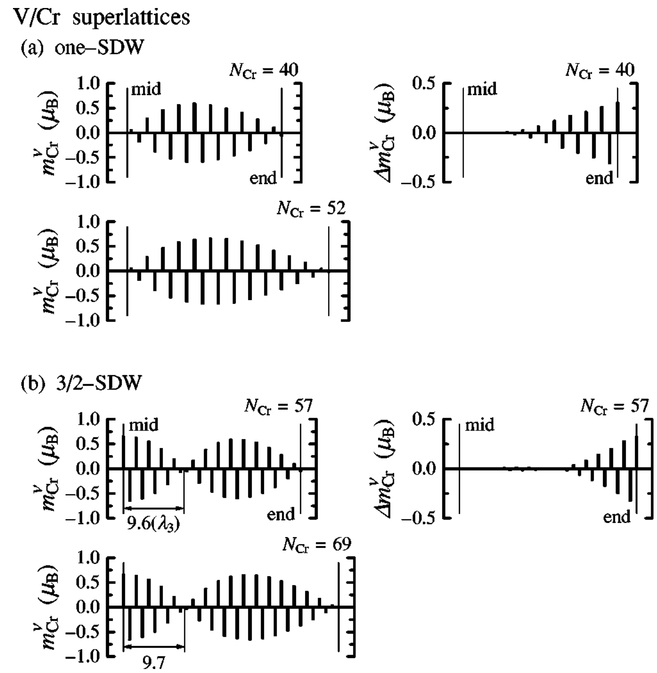

FIG. 4. Change of the (a) one SDW and (b) 3/2 SDW orders in the V/Cr superlattices, where a right half of the distribution of the Cr local magnetic moments m^{ν}_{Cr} is shown for $N_{Cr}=40,52$ and $N_{Cr}=57,69$, respectively, and a value under an arrow indicates λ_3. Difference of the distributions Δm^{ν}_{Cr} from that of $N_{Cr}=52$ (69) is also shown for the one SDW (3/2 SDW) order.

node around the quarter position λ_2, double of which corresponds to the period of SDW, becomes a constant of 20 monolayers (see the lower right of Fig. 3), and the largest magnitude between the middle and the node becomes a constant of $0.67\mu_B$. The difference of the distributions Δm^{ν}_{Cr} between $N_{Cr}=68$ and 76 is fairly small in comparison with that between $N_{Cr}=60$ and 76, and the distributions for larger N_{Cr}'s become alike to each other in the vicinity of the middle of the Cr layer; the change of the 3/2 SDW order takes place at around 68.

Thus the one SDW or 3/2 SDW order in the vicinity of the middle of the Cr layer is roughly insensitive to N_{Cr} and resembles the SDW in bulk Cr, with regard to its period and amplitude, for a larger N_{Cr}. The order, however, is roughly insensitive to N_{Cr} around the interface at the same time, as can be noticed in Figs. 2 and 3, which means that the order still undergoes large influence of the Fe layers around the interfaces. It can be concluded that the modulation of the one SDW or 3/2 SDW order is given rise to by competition between an SDW controlled by the Fe layers and an SDW growing spontaneously in the Cr layer itself.

On the other hand, the SDW order in the V/Cr superlattices offers a marked contrast to that in the Fe/Cr one. In particular, there are not antinodes but nodes around the interfaces between the V and Cr layers.[11] This is because the V layer is magnetically less significant than the Fe one and a smaller magnitude of the Cr local magnetic moment is favored around the interface. Solutions of the half SDW, one SDW, and 3/2 SDW orders appear for odd $N_{Cr}\geqslant 13$, even $N_{Cr}\geqslant 24$, and odd $N_{Cr}\geqslant 37$, respectively, which correspondence to the parity of N_{Cr} is just the reverse of the Fe/Cr superlattices. For the half SDW order, existence of the critical thickness for its appearance suggests that the SDW order in the V/Cr superlattices may grow spontaneously, but we cannot find a sign of the SDW in bulk Cr.

The sign can be found for the one SDW and 3/2 SDW orders, as can be seen in Fig. 4, where the distribution of the Cr local magnetic moments is shown with N_{Cr} varying. For the one SDW order, the distribution itself does not display as evident a sign as the variation of λ_1 for the one SDW order in the Fe/Cr superlattices, but the difference of the distributions Δm_{Cr}^{ν} shows that the distributions are alike to each other in the vicinity of the middle of the Cr layer. This is also true for the 3/2 SDW order, and furthermore it is found that an interval from the middle to the node λ_3, four times of which corresponds to the period of SDW, is around 10 monolayers and $|m_{Cr}^{mid}|$ is around 0.66 μ_B. The one SDW order with $N_{Cr}=40$ or 3/2 SDW order with $N_{Cr}=57$ resembles the SDW in bulk Cr as a whole, and the distribution in the vicinity of the middle of the Cr layer is roughly unchanged against an increase of N_{Cr}.

Thus the one SDW or 3/2 SDW order in the vicinity of the middle of the Cr layer resembles the SDW in bulk Cr, except for the case where the Cr layer is not thick enough to contain one or 3/2 period of the SDW in bulk Cr. This is consistent with the notion that the SDW order in the V/Cr superlattices grows spontaneously. It is, however, to be noted that influence of the V layers is rather large and not so simple as expected; the nodes never leave the interface toward the inner Cr layer, and the SDW order undergoes considerable modulation even for the inner Cr layer far away from the interface.[12] Although the V layers is magnetically less significant, the proximity effects of the V layer are substantial.

In conclusion, the SDW order in the Fe/Cr and V/Cr superlattices grows spontaneously and resembles the SDW in bulk Cr, in the vicinity of the middle of the Cr layer, for a sufficiently large N_{Cr}. A clear crossover from the SDW order governed by the proximity layers to the spontaneous SDW order is found in the Fe/Cr superlattices, where the critical thicknesses of the crossover are estimated at around 49 and 68 for the one SDW and 3/2 SDW order, respectively. The ferromagnetic Fe or nonmagnetic V boundary layers still have some influence upon the SDW orders around the interfaces, nevertheless, even for such a large N_{Cr}. The influence may extend to 10 monolayers or more from the interface for both the Fe/Cr and V/Cr superlattices, though analysis of the proximity effects of the boundary layer is not so straightforward; the analysis is an issue of the future study, where discrimination between the bulk and proximity effects on the SDW order will be discussed, together with analysis of the electronic structure. Last, it is noted that the self-consistent solution of the SDW order discussed here does not necessarily mean a stable one but may be a metastable one, since the stable solution is determined together with the calculation for the case of antiparallel magnetizations; some of the spontaneous SDW orders, however, are surely stable for certain N_{Cr}'s.

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