k-Space Origin of the Long-Period Oscillation in Fe/Cr Multilayers: A Photoemission Study of Epitaxial Cr Grown on an Fe(100) Whisker

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The *k*-space origin of the long-period (18 Å) oscillation in the interlayer magnetic coupling of Fe/Cr(100) multilayers is investigated by means of angle-resolved photoemission by probing quantum well (QW) states at the Fermi energy for epitaxial Cr grown on an Fe(100) whisker at 300 °C. The periodicity of the intensity oscillations of the Cr QW states in the vicinity of the *d*-derived "lens" feature of the Fermi surface is 17 ± 2 Å. Thus the lens is identified as a prime candidate for the origin of the long period. [S0031-9007(96)02272-7]

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The oscillatory interlayer magnetic coupling in giant magnetoresistance (GMR) multilayers is mediated by the energetics of magnetic quantum well (QW) states in the spacer [1–3]. Electron spectroscopies have been used to probe these states in overlayers on magnetic substrates [1,4–7]. The thickness periodicity of these spin-polarized QW states to cross E_F coincides with that of antiferromagnetic coupling [1,4]. The QW states are confined by the spin-dependent band offsets of the magnetic layer [4,8], which determine the strength of the coupling [9], while its *periodicity* is determined by extremal spanning vectors of the spacer's Fermi surface [10], as in the Ruderman-Kittel-Kasuya-Yosida (RKKY) coupling model [11]. In the present work we focus on the latter aspect and explore the *k*-space origin of the long-period (18 Å) oscillation of Fe/Cr(100) multilayers by monitoring the emergence of QW states in Cr overlayers on an Fe(100) whisker. This is of interest because of the controversies surrounding the long period of Fe/Cr. While the short period is universally recognized as emerging from the nested feature of the Fermi surface, which also gives rise to the antiferromagnetism of bulk Cr, the origin of the long period remains elusive. This, taken with the following facts, heightens interest in the problem: (i) Fe/Cr is a prototypical GMR material [12–14], (ii) most other GMR materials have a shorter long period of only ~12 Å, and (iii) transition-metal spacers like Cr have more complex and challenging Fermi surfaces than the Cu of the Co/Cu prototype most studied to date theoretically and spectroscopically.

There are several regions of the Cr Fermi surface whose spanning vectors are candidates to explain the 18-Å long period. Koelling [15] suggested that the relatively isotropic *d*-derived "lens" (*a* in Fig. 1) provides the long-period caliper. Van Schilfgaarde and Harrison [11] attribute it to aliasing, whereby discrete sampling of the short period (*b* in Fig. 1) at lattice sites yields the long period. Mirbt *et al.* [12] proposed that the relevant spanning vector is at the Brillouin zone (BZ) center and is accompanied by a symmetry breaking to a CsCl structure (not marked). Stiles [13] argued that the caliper of the *sp*-derived *N*-centered ellipse (*c* in Fig. 1) provides the long-period coupling. Herein we monitor the appearance of QW states near E_F that have the same $k_\parallel$ as these spanning vectors and the periodicity of their intensity oscillations at E_F as a function of Cr thickness of Fe(100). We find that the intensity at E_F oscillates with a period

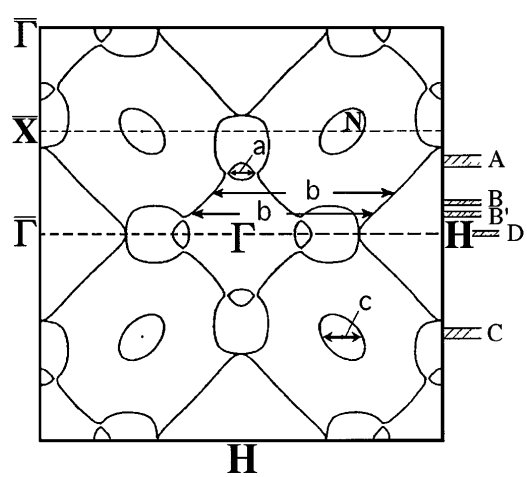

FIG. 1. Calculated Fermi surface of bulk Cr [11]. The high-symmetry points of the surface BZ are indicated by the dashed lines and on the left. The extremal spanning vectors of the interest are marked with *a*, *b*, *c*, and the experimental $k_\parallel$ at E_F are marked with *A*, *B*, *B'*, *C* and *D*, as discussed in the text.

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of 17 Å in the vicinity of the lens (*a* in Fig. 1), while it oscillates, if at all, at 2.6 Å at the nesting region (*b* in Fig. 1). This rules out aliasing as the origin of the long period [11] and supports the theoretical identification of the *d*-derived lens as the region of interest [15].

The experiment is challenging for a number of reasons: (i) the relatively flat *d* bands leave only very narrow energy windows near E_F for QW states to exist and to shift with film thickness [10]; (ii) the investigation of QW states away from the zone center is demanding [15]; (iii) there are multiple bands near E_F, which produce high backgrounds underlying the QW-induced intensity oscillations; and (iv) prerequisite for the identification of QW states is an atomically smooth and flat surface. Room-temperature (RT) growth of Cr on Fe(100) is known to result in relatively rough films, thus we used an Fe whisker substrate which is known to be atomically smooth and followed the 300 °C growth procedure of Ref. [16]. The Fe(100) whisker was cleaned *in situ* with cycles of Ne sputtering at 25–600 °C and annealing at 700 °C. Cr was deposited at a rate of 0.05–0.3 Å/min, which was monitored with a quadrupole mass spectrometer and calibrated with the photoemission intensities of the Cr and Fe 3*p* levels, assuming an effective mean free path of 5Å. The accuracy of the absolute thicknesses is estimated as within 30%, while the relative thickness in each deposition sequence is more reliable due to the stability of the evaporators. Films grown at this condition are known to grow in a nearly perfect layer-by-layer mode, though the first 5–6 Å form an alloy with Fe [17]. In this work, films of 8–50 Å are discussed to avoid the alloy region. Both the substrate and the films are well ordered as indicated by 1×1 LEED patterns with the extremely sharp spots characteristic of Fe whiskers, indicating superior epitaxial growth of Cr. The chamber pressure during both the depositions and measurements was in the low 10^{-11} Torr range. No impurity peaks were detected with photoemission for the substrate and initial Cr growth.

Angle-resolved photoemission was carried out at the U5 undulator beam line at the National Synchrotron Light Source, Brookhaven National Laboratory. The spectra were mainly taken at 59.7 eV to probe the *d*-derived states, with some at 31.8 eV for comparisons. The energy resolution was ~0.15 eV and the angular resolution was ~1°. The incident angle of the light was 35° with respect to the surface normal. The photoelectron parallel momentum, and therefore the corresponding wave vector $k_{\parallel}$, was varied by moving the analyzer away from the sample normal direction in the incident plane. The photoemission intensities are normalized to the background intensity at high binding energy. The thickness dependence was measured by sequentially depositing more Cr before each set of measurements. All measurements were made at ambient temperature.

Figure 2 shows the thickness dependence at one part of the Cr BZ with an emission angle of 12°, as marked *A* at E_F on Fig. 1. The peak at 0.95 eV binding energy (E_b)

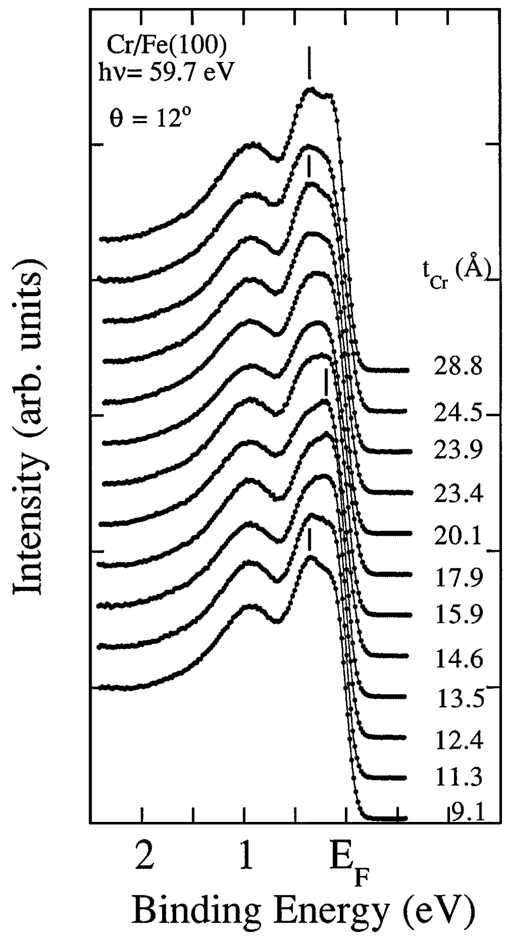

FIG. 2. Spectra of epitaxial Cr films on Fe(100) at $h\nu =$ 59.7 eV and $\theta = 12°$.

remains constant both in binding energy and intensity throughout the whole thickness range. It is a known surface state [18] and can serve as a reference for the thickness-dependent QW states. In the region of $E_b =$ 0–0.5 eV, the relative intensities of the peaks vary with film thickness in a nonmonotonic fashion. Although it is difficult to resolve the peaks for all the thicknesses possibly due to limited resolution, the curve fitting of the spectra reveals that the E_b of the peak at ~0.3 eV shifts with film thickness. For fixed thicknesses, the peak exhibits no dispersion with perpendicular momentum when $h\nu$ is changed from 59.7 to 31.8 eV, though its intensity decreases. This suggests that the state is two dimensional (2D) and *d*-like. We therefore attribute the relative intensity changes to the thickness-dependent *d*-QW states that shift between 0–0.5 eV.

Figure 3 shows the intensity variation with the film thickness at E_F, as determined from the intensity at the midpoint of the Fermi edge. $k_{\parallel}$ is given, in units of $Å^{-1}$, as

$$k_{\parallel} = 0.512\sqrt{h\nu - E_b - \phi}\,\sin\theta\,,$$

where ϕ is the work function of Cr(100) (4.5 eV) and θ is the electron emission angle from the surface normal.

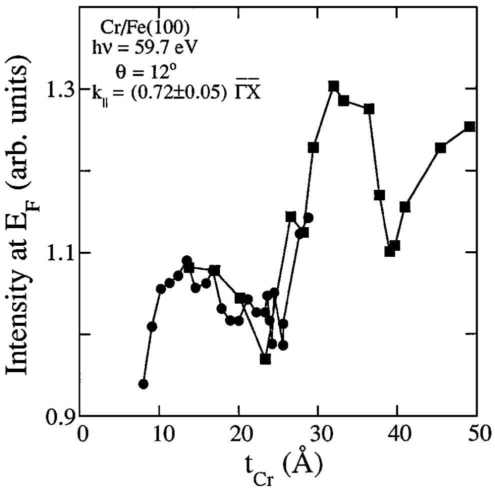

FIG. 3. The intensities at E_F vs film thickness at $k_{\|} = 0.72$ in units of the surface BZ, $\overline{\Gamma}\,\overline{X}$ (around the lens, *A* in Fig. 1), which yield a periodicity of 17 ± 2 Å. Independent thickness sequences are indicated with different symbols.

At 12°, $k_{\|}(E_F) = 0.79 \pm 0.05$ Å^{-1}, i.e., $0.72 \pm 0.05\overline{\Gamma}\,\overline{X}$ in units of the surface BZ, using the Cr lattice parameter of 2.88 Å. The surface BZ boundary $\overline{X}$ is experimentally determined as 1.15 ± 0.10 Å^{-1} from the 2D band dispersions, consistent with the calculated value of 1.09 Å^{-1}. $k_{\|}(E_F)$ for the 12° spectra is close to that of the *d*-derived lens feature located at 0.6–0.7 $\overline{\Gamma}\,\overline{X}$ according to different calculations [10,11,19]. Two independent thickness sequences are plotted in Fig. 3 with different symbols. It is apparent that the intensity at E_F possesses an oscillatory behavior with a period of 17 ± 2 Å, i.e., 12 ± 1 ML. This oscillation period coincides with the 18-Å long period of the magnetic coupling through Cr [20]. The oscillatory thickness dependence of the intensity indicates the existence of QW states that cross E_F periodically with film thickness. Our results, therefore, identify the vector across this *d*-derived lens (*a* in Fig. 1) or the neck just outside of it as a viable candidate for the origin of the long period.

To verify our methodology, we performed similar measurements during the same sets of depositions at 5° off normal, where the nesting vector resides (marked *B* in Fig. 1). As in Fig. 4(a), the intensities of the peaks vary in a subtle but systematic manner when observed on close inspection. For instance, the intensity at 0.3 eV is nonmonotonic with film thickness. This variation is evident in Fig. 4(b) after subtraction of a linear background. Similar to the QW state observed at $\theta = 12°$, the state at ~0.3 eV is also 2D in character (no dispersion with $h\nu$). The intensity at E_F for the spectra at 5° ($k_{\|} = 0.33 \pm 0.02$ Å or $0.30 \pm 0.02\ \overline{\Gamma}\,\overline{X}$) are shown in Fig. 5. The difference in behavior between the 5° and 12° spectra for the same deposition indicates that the intensity variations in Fig. 3 are not accidental and suggests that our methodology is feasible. While

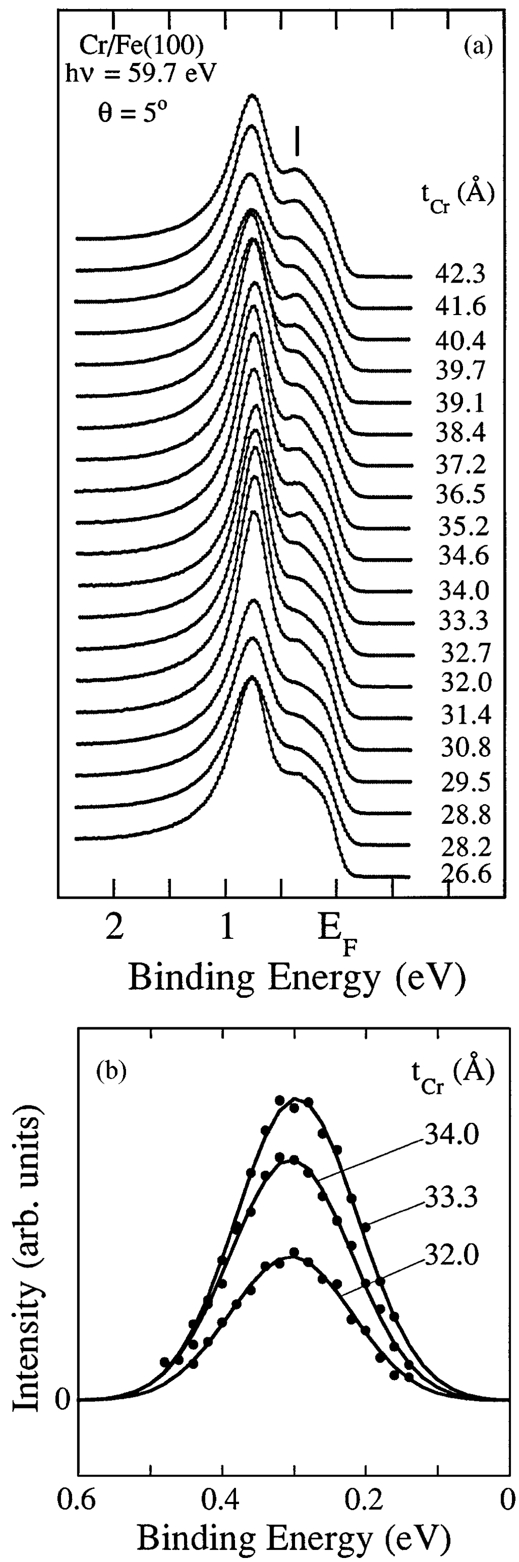

FIG. 4. Spectra of epitaxial Cr films on Fe(100) at $h\nu = 59.7$ eV and $\theta = 5°$. (a) Original spectra normalized to the background intensity; (b) after subtracting a linear background.

it is not clear if the scatter of several data points is related to aliasing [21] or simply noise, it is evident that, unlike in Fig. 3, there is no well-defined 18-Å long period

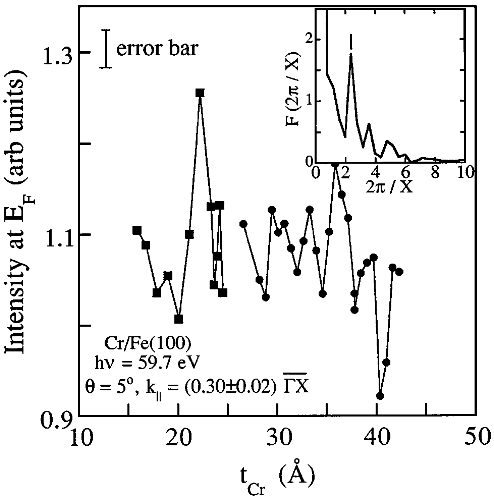

FIG. 5. The intensities at E_F vs film thickness at $k_\parallel = 0.30$ (the nesting region, B in Fig. 1), in units of the surface BZ, $\overline{\Gamma}\,\overline{X}$. Independent thickness sequences are indicated with different symbols. The inset is the Fourier transform which yields a periodicity of 1.8 ± 0.3 ML.

in Fig. 5. This rules out the long-period oscillation as arising from aliasing at the nesting region. In fact, the aliasing mechanism has been dismissed independently by a scanning electron microscopy with polarization analysis (SEMPA) experiment [21] since it possesses a different periodicity. A Fourier transform of the data in Fig. 5, shown in the inset, indicates a peak at 2.4 $Å^{-1}$, that is, a period of 2.6 ± 0.4 Å (1.8 ± 0.3 ML). This short-period oscillation is less clear than the long-period one probably due to its smaller amplitude and the enhanced influence of any random errors in the film thickness. The *ratio* of the long and short periods of the intensity oscillations at E_F, independent of any systematic error in thickness calibration, is 6.5 ± 1.2, the same as the 6.25 ratio of the long-to-short period of the oscillatory magnetic coupling (18 Å [20] and 2.88 Å [16]). This supports our argument that the long-period oscillation originates from around the *d*-derived lens (*a* in Fig. 1) of the Cr Fermi surface and is consistent with the accepted interpretation that the short period originates from the nesting. Incidentally, this assignment is consistent with the fact that the long period for Fe/Cr(100) and Fe/Cr(211) superlattices are the same [22], since the lens is a relatively isotropic feature of the Fermi surface [15].

It should be noted that as a methodology to conclusively correlate a particular part of the Fermi surface to the coupling one should search over the entire 2D BZ since multiple periodicities can exist. Data at normal emission (D in Fig. 1) do not reveal additional peaks, though the intensity of the peak at 0.67 eV varies (10–20)% with a short period of 3.2 ± 0.5 Å with film thickness. This indicates that the long-period does not originate from the zone center. Another potentially interesting area is the ellipse centered around the N point, where the states have *sp* character. Limited results at $h\nu = 31.8$ eV (C in Fig. 1), however, have not revealed new states near E_F, and the change in intensity at E_F is only about 2% in the range of 26–32 Å, similar to the error bar of our results for 5° and 12° at $h\nu = 59.7$ eV. Further studies are warranted at low photon energies in this part of the BZ.

In conclusion, we observe *d*-QW states in Cr near E_F that possess the periodicity of the oscillatory interlayer magnetic coupling. This identifies the *d*-derived lens of the Fermi surface as a region of prime interest for the long-period coupling oscillation, although a more complete *k*-space sampling should be fruitful to further evaluate the relationships between QW states and magnetic coupling. Our work demonstrates that angle-resolved photoemission provides a novel methodology to distinguish the *k*-space origins of different oscillation periodicities in the interlayer magnetic coupling. This can, in principle, be broadly utilized in studies of the electronic structure of GMR multilayers.

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