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Magnetism and interlayer coupling in fcc Fe/Co films

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The magnetism of epitaxial fcc Fe films deposited on Co(100) and sandwiched between two Co(100) films was investigated by x-ray magnetic circular dichroism. The dependence of the Fe magnetism on the film thickness is complex and qualitatively similar on Co(100) and in fcc Co/Fe/Co(100) trilayers. The fcc Fe film magnetization presents a pronounced oscillation, suggesting a partial antiferromagnetic ordering in the 5–10 monolayer thickness range. The fcc Fe films mediate an oscillatory, indirect coupling in Co/Fe/Co(100) structures that alternates in correspondence with the changes of the Fe magnetization.

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I. INTRODUCTION

Fe is probably the most extensively studied magnetic element, but it still represents a challenging system for experimental and theoretical research. In particular, the fcc Fe phase attracts a lot of attention because of the complex interdependence between its structural and magnetic properties.[1–9] In contrast to bcc Fe which is a ferromagnet with a large moment ($>2.2\mu_B$/atom), fcc Fe is theoretically expected to order in a low-spin antiferromagnetic state at the equilibrium lattice constant. Electronic band structure calculations predict that a small compression or expansion of the fcc lattice gives rise to magnetovolume instabilities converting the low-spin antiferromagnetic state into a nonmagnetic or a high-spin ferromagnetic state, respectively.[1] In addition, recent total energy calculations indicate that various noncollinear configurations might be favored in certain ranges of structural parameters.[10]

The magnetism of bulk fcc Fe is hardly accessible to the experiment since the fcc phase is only stable at temperatures above 1183 K. Metastable fcc Fe films, however, grow epitaxially on the fcc(100) surfaces of the late $3d$ transition metals (Co, Ni, Cu) and significantly simplify the experimental study of the magnetic properties. On all of these substrates fcc Fe films display a similar interplay between structural and magnetic properties.[6] Films thinner than 5 monolayers (ML) have a slightly expanded atomic volume due to a tetragonal distortion. They order magnetically in a high-spin ferromagnetic structure with a nearly uniform distribution of large magnetic moments ($>2\mu_B$/atom). Fe films of larger thickness (5–10 ML) exhibit a relaxed, almost isotropic fcc lattice and an enhanced interlayer distance between surface and subsurface layers. The magnetization of the films in this thickness range is significantly reduced ($<1\mu_B$/atom). Above 10 ML Fe film thickness, the fcc structure becomes unstable and gradually converts to the bcc lattice of bulk Fe.

Despite extensive efforts the magnetic structure of fcc Fe films in the 5–10 ML thickness range has not been fully clarified. The small magnetic signal that persists in this range is attributed to Fe moments located at the film surface, subsurface, and buried interface with the substrate. Various studies show that the free Fe surface is ferromagnetically ordered and give some indication for a ferromagnetic coupling to the subsurface layer.[2,3,8,9,11] The magnetic ordering of the interface layer, instead, is controversial.[6,9,11] X-ray magnetic circular dichroism (XMCD) gives indications for Fe ferromagnetism at the fcc Fe/Co(100) interface, whereas recent Kerr and photoemission studies conclude that only the Fe surface is ferromagnetically ordered.

The inner Fe layers are generally believed to be paramagnetic or antiferromagnetic[3–5] and therefore not to contribute to the total magnetic moment. No evidence for antiferromagnetism in fcc Fe films on Co(100) has been reported until now. Mössbauer and magneto-optical Kerr effect (MOKE) measurements indicate instead some kind of antiferromagnetic arrangement in fcc Fe on Cu(100) at low temperatures ($T<200$ K). Such a magnetic ordering of the films should also influence the magnetic properties of multilayers containing fcc Fe films. A study of Co/Fe/Co(100) trilayers reports antiparallel coupling for all Fe thicknesses in the 5–10 ML range.[12] The microscopic relation between the interlayer coupling and the magnetic structure of the fcc Fe films has not yet been established.

This work examines the magnetic properties of fcc Fe films grown on Co(100) and of fcc Co/Fe/Co(100) trilayers. XMCD measurements show that the magnetism of Fe films on Co(100) is more complex than reported in previous studies. The Fe magnetization is large up to 5 ML thickness and it is further enhanced in the monolayer regime. The magnetic signal of 5–10 ML Fe films is significantly weaker. In addition, it exhibits a pronounced dependence on the film thickness. The magnetization of the fcc Fe films remains qualitatively similar in Co/Fe/Co(100) trilayers. The fcc Fe mediates an oscillatory coupling between the Co layers, with a clear correlation between the sign of the interlayer coupling and the magnetization of the fcc Fe films.

II. EXPERIMENT

X-ray magnetic circular dichroism was used to investigate the magnetism of Fe films on Co(100) and of Co/Fe/Co(100) epitaxial trilayer structures. Epitaxial Fe and Co(100) layers were deposited onto a Cu(100) substrate. The Cu substrate was prepared by repeated cycles of ion sputtering and annealing until it displayed sharp low-energy electron diffraction $p(1\times1)$ spots. The clean and well-ordered Cu(100) surface was covered with thin epitaxial Co(100) films. Onto these Co films, Fe films (1–11 ML) were deposited in a wedge geometry (1 ML/mm slope) to ensure identical growth conditions for films of different thicknesses. The base pressure was below 1×10^{-10} mbar and did not rise above 6×10^{-10} mbar during the evaporations. The thickness calibration was performed by means of a quartz crystal microbalance. During the film growth and the x-ray absorption measurements the sample was kept at room temperature, unless specifically mentioned.

The dichroism measurements were performed with circularly polarized radiation at the PM3 beamline at BESSY I. The light incidence angle with respect to the surface normal was 39°. A magnetic field pulse was applied along the [110] in-plane direction to remanently magnetize the sample. Dichroism effects in the absorption spectra were determined by reversing the magnetization direction. Fe and Co $L_{2,3}$ absorption spectra were measured by total electron yield. Figure 1(a) illustrates as an example the $L_{2,3}$ absorption spectra for 3.1 ML and 5.3 ML Fe films grown on an 8 ML Co film. The figure shows the original spectra without corrections for the incomplete circular polarization of the light ($\approx 80\%$) and for the angle between the light polarization and the sample magnetization. The difference between the spectra for the two magnetization directions (R and L) gives the dichroism spectrum (R-L).

In the following, no attempt will be performed to derive absolute values for the magnetic moments through the application of sum rules. Note that multiplet effects might be important for the determination of the magnetic moments.[13] As a measurement of the dichroism signal we use the relation

$$I \sim \frac{L_3^L - L_2^L}{L_3^L + L_2^L} - \frac{L_3^R - L_2^R}{L_3^R + L_2^R},$$

where $L_{3[2]}^{R[L]}$ represents the maximum of the L_3 [L_2] edge for parallel (R) [antiparallel (L)] alignment of light polarization and magnetization direction, after a linear background subtraction. This dichroic signal is proportional to the magnetic moment and it monitors the variations of the dichroic effect in a reliable and sensitive way. These measurements yield element-specific information on the averaged magnetic moments (i.e., the magnetization) along the direction of the light polarization. The different magnetic layers contribute to the total signal with a weight that decreases exponentially with the distance from the surface with an attenuation length of about 15–20 ML.

III. RESULTS AND DISCUSSION

A. Fe on Co(100)

Figure 1(b) shows the dichroism signal for Fe films on Co(100). In agreement with earlier investigations the Fe dichroic signal is large for thicknesses up to 4 ML and it is strongly reduced for 5–10 ML film thickness.[6,9] The Fe dichroic signal increases again as the Fe films gradually convert into the bcc structure (>10 ML), forming (110) domains oriented into four different directions.[14] The Fe growth does not significantly affect the Co dichroism signal up to a film thickness of 10 ML. Above this thickness the Co dichroic signal decreases, possibly because the interface coupling tends to align also the Co moments along the [100] easy axis of the bcc Fe domains.

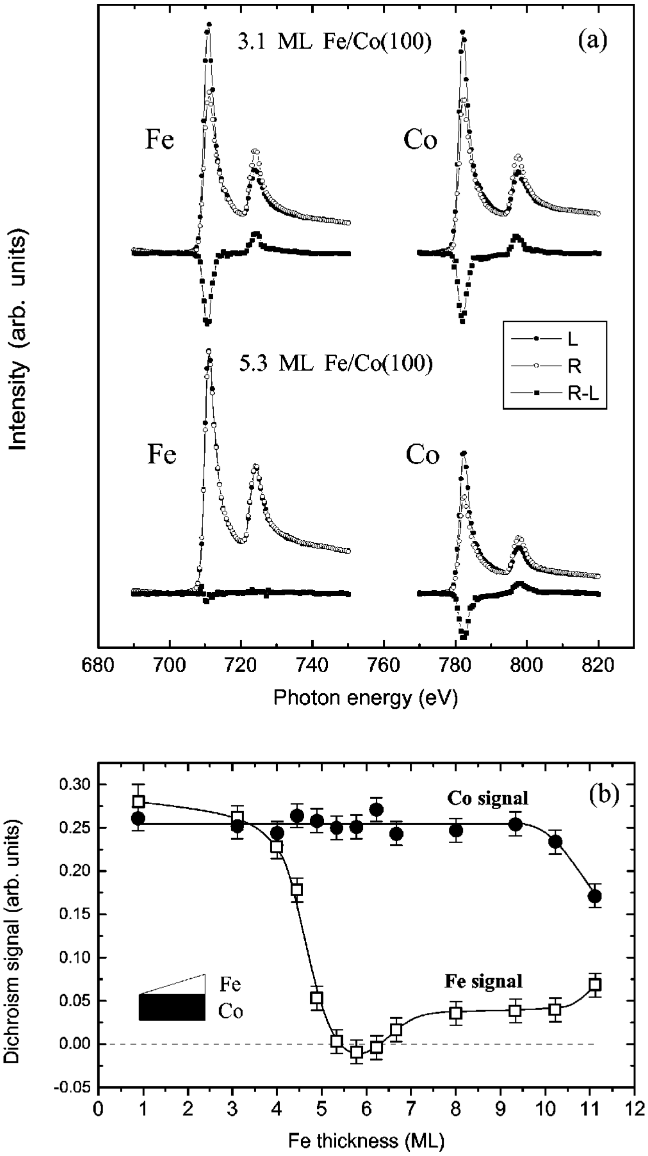

FIG. 1. (a) Fe and Co x-ray absorption spectra for parallel (R) and antiparallel (L) alignment of light polarization and magnetization direction and the corresponding dichroism spectra (R-L) determined for 3.1 ML and 5.3 ML Fe on 8 ML Co(100). Note that these are original data that are not corrected for the angle between light polarization and sample magnetization or for the incomplete circular light polarization. (b) Fe and Co dichroism signals (open squares and solid circles, respectively) determined for the Fe/Co(100) system as a function of Fe film thickness. Note that the solid lines, as well as the dashed lines in Figs. 2 and 3, are only meant as a guide to the eye.

The largest Fe magnetic signal is found at the smallest examined Fe film thickness of 0.9 ML. Large Fe moments in the monolayer regime are not surprising and can be ascribed to the bonding with the Co substrate as well as to the low coordination of the Fe atoms. The bonding with Co increases the occupation of the Fe 3d spin-up states and enhances the Fe moment in Co-rich Fe-Co alloys [(2.6–3.0 μ_B) (Refs. 15 and 16)] and at the bcc Fe/Co(100) interface [(2.5–2.6 μ_B) (Ref. 17)] in comparison with bcc Fe metal. The bonding at the fcc Fe/Co interface is likely to induce a similar effect. The low atomic coordination at surfaces and in monolayers can also significantly enhance the Fe moment.[16] With increasing film thickness (<4 ML) the magnetic signal slightly decreases, as the relative contribution from the interface and surface layers becomes smaller.

In 5–10 ML films the Fe magnetization is much weaker than in the thinner films. It displays a pronounced minimum and vanishes at about 6 ML thickness. The minimum is followed by a positive and nearly constant signal between 7 ML and 10 ML. This complex behavior has not been observed in previous investigations. XMCD and MOKE studies on Fe/Co(100) (Refs. 6, 9, and 12) report instead over the whole 5–10 ML thickness range a nearly constant or monotonously decreasing magnetic signal. The magnetic signal has been attributed to the ferromagnetism of the surface (or interface), whose magnitude and orientation was assumed to be independent of the film thickness.

The observation of a pronounced structure in the magnetization of Fe films of 5–10 ML thickness has some direct implications on the magnetism of the films. A simple magnetic configuration where only the surface (or the interface layer) is ferromagnetically ordered does not explain the zero magnetization for 6 ML films and can therefore not describe the whole 5–10 ML thickness range. Also, the pronounced minimum observed in the Fe magnetization curve excludes a ferromagnetic configuration with uniformly distributed, low moments in the films. The modulation of the Fe magnetization rather suggests that antiferromagnetic order in parts of the films leads to a cancellation of the moments to an extent that depends on the film thickness.

The simplest antiferromagnetic structure one can consider is layerwise antiferromagnetic [i.e., a stack of ferromagnetic Fe(100) monolayers with magnetization alternating in opposite direction]. This magnetic structure would produce an oscillatory behavior of the film magnetization with a 2 ML period, with minima and maxima for even and odd numbers of monolayers, respectively. This structure could account for the minimum observed at 6 ML thickness, but not for the low signal at 5 ML and the nearly constant signal for Fe films thicker than 7 ML.

Recent calculations predict that a number of nearly degenerate states can be obtained by varying the stacking sequences of ferromagnetic monolayers in fcc Fe films on Cu(100).[7] An oscillatory MOKE signal from fcc Fe on Cu(100) was observed by Li *et al.* for temperatures below 200 K.[3] The MOKE signal of Fe/Cu(100) exhibits minima with a separation of 2.6 ML, suggesting that the Fe films order in an incommensurate spin-density wave rather than in a simple layerwise antiferromagnetic structure. The authors pointed out, however, that Kerr effect modulation might also reflect changes of the electronic structure due to electron confinement in the films that would not be related to the magnetic moments in a simple way.

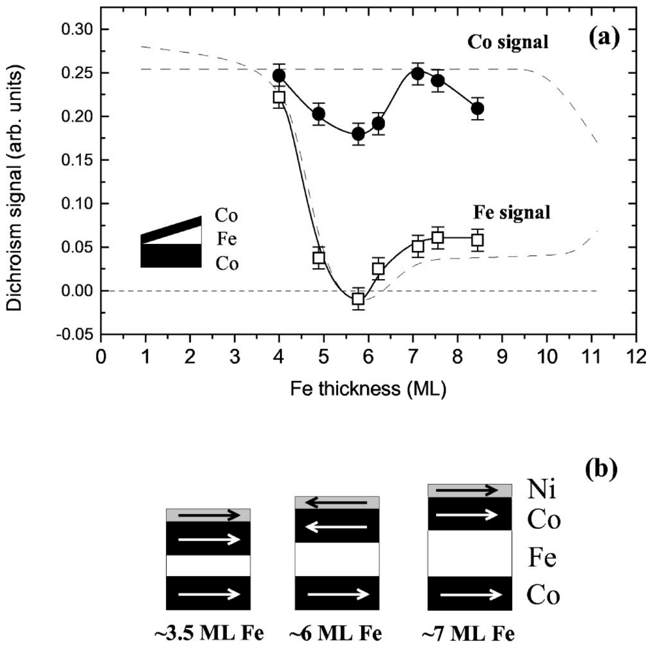

FIG. 2. (a) Fe and Co dichroism signals (open squares and solid circles, respectively) determined for the trilayer 2 ML Co/Fe/8 ML Co(100) system as a function of Fe film thickness in comparison with the results of the Fe/8 ML Co(100) system (dashed lines). (b) Schematic picture for the magnetic configuration in a Ni/Co/Fe/Co(100) system for 3.5, 6, and 7 ML Fe. The Ni film on top of the Co/Fe/Co sandwich directly probes the magnetization direction of the upper Co film.

The present XMCD results unambiguously prove that the magnetic moments in the fcc Fe films change in a noncontinuous way in the 5–10 ML range. Although it is definitely not possible to completely derive the magnetic structure of the Fe films from the present data, the results suggest that the inner layers might be antiferromagnetically ordered already at room temperature. Proximity effects can explain the enhanced ordering temperature in Fe/Co(100) with respect to the Fe films on Cu(100). In comparison with Fe/Cu(100), additional complexity in the Fe/Co(100) system might arise from the exchange interactions at the interface. In particular, the unavoidable presence of steps at the substrate surface might frustrate layerwise antiferromagnetic structures in fcc Fe films.

B. Co/Fe/Co(100)

In view of the apparent complexity of the Fe magnetic behavior, it is surprising that it is qualitatively maintained in Co/Fe/Co(100) trilayers. Figure 2(a) compares the Fe dichroism signal for Fe/8 ML Co(100) and for a 2 ML Co/Fe/8 ML Co(100) trilayer. The development of the Fe dichroism signal in films of 4–9 ML thickness remains strikingly similar despite the additional Co film. While other works associated the moment of fcc Fe films on Co(100) thicker than 5 ML exclusively with the ferromagnetism of the free Fe surface,[11] the present results show that also sandwiched Fe films have a nonzero total magnetic moment. This again indicates that the

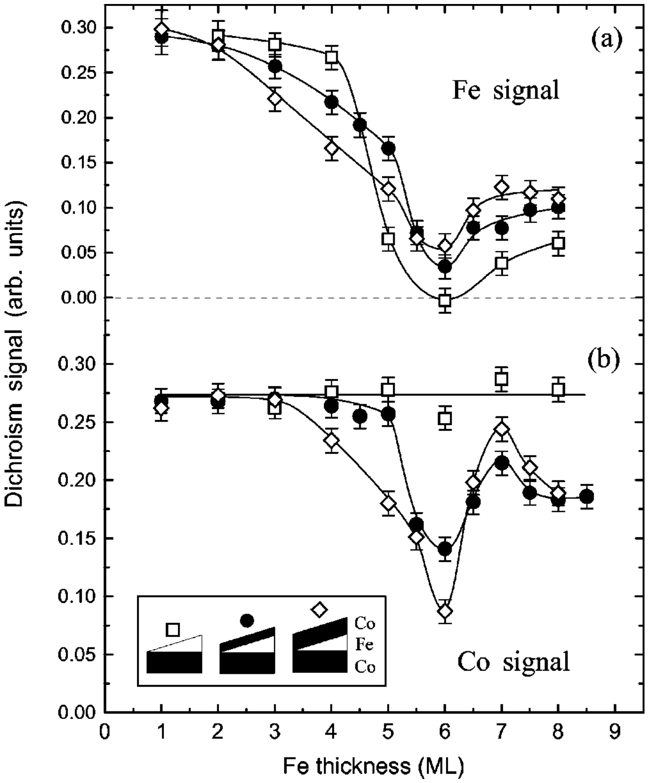

FIG. 3. (a) Fe dichroism signals determined for an uncovered Fe film on 20 ML Co (open squares) and for the same sample covered with 4 ML Co (solid circles) and 8 ML Co (open diamonds). (b) Co dichroism signals for the same systems as shown in (a).

fcc Fe film magnetization does not only derive from the surface. In fact, the Co-covered films of thicknesses above 6 ML exhibit a slightly larger Fe signal than the uncovered ones, possibly because the coupling with the top Co film increases the ferromagnetic order in the Fe interface layer.

The Co magnetization shows pronounced changes in the Co/Fe/Co(100) trilayers as a function of Fe thickness. The Co dichroism signal of the 2 ML Co/Fe/8 ML Co(100) trilayer presents an oscillatory behavior [Fig. 2(a)]. As both the Co substrate and Co top layer contribute to the total dichroic intensity with different weights reflecting the film thickness and the probing depth of the measurement, the reduced signal indicates that the two layers are not aligned in parallel. This is indeed confirmed by the more pronounced signal oscillations observed when increasing the thickness of the top Co layer. Figures 3(a) and 3(b) show the Fe and Co dichroism signals for 4 ML Co/Fe/20 ML Co and 8 ML Co/Fe/20 ML Co trilayers. The Co signal displays similar structures in these trilayers as for the 2 ML Co/Fe/8 ML Co(100). Assuming an escape depth of about 20 ML, the Co signal for 6 ML Fe can be accounted for by a nearly antiparallel orientation of the moments in the two Co films. The main additional effect of thicker Co films is that the transition from large to small Fe magnetization becomes increasingly broader. This suggests that the tetragonally distorted structure of the uncovered Fe films partially relaxes in the trilayers into the isotropic fcc structure with low magnetization. Evidence for the metastable character of the tetragonal distortion of 4 ML Fe/Cu(100) films has been reported previously.[18] Figure 3(a) also clearly shows that the Fe signal in the regime of reduced magnetization increases after the deposition of 4 ML and 8 ML Co.

In order to directly probe the orientation of the moments in the top Co film with respect to the bottom Co layer, a Ni monolayer was deposited on the 2 ML Co/Fe/8 ML Co(100) trilayer. The Ni and Co moments are expected to couple parallel. Therefore, the Ni layer probes the magnetization direction of the upper Co film. The magnetization of the thicker bottom Co layer can be assumed to remain locked along the easy axis direction. The sign of the Ni dichroic signal indicates a predominant parallel coupling between the two Co films at 3.5 ML Fe, antiparallel for 6 ML Fe and again parallel for 7 ML Fe as illustrated in Fig. 2(b). This result thus proves that fcc Fe mediates an indirect coupling between the Co films with a fast oscillatory behavior. To our knowledge, this is the first experimental evidence for an oscillatory coupling in a system consisting of two elements that are both ferromagnetic in their normal structure.

There appears to be a clear correspondence between the Fe magnetization and the coupling that it mediates between the Co layers. The coupling is positive across the thin ferromagnetic Fe films; it switches to negative in correspondence with the magnetization minimum at 6 ML and again to positive at 7 ML Fe thickness. The correspondence between the oscillatory behavior of the coupling and the Fe magnetic signal indicates that the magnetic order of the Fe films has a decisive role in determining the coupling. In this respect, it should be noticed that the Fe and the Co signals do not show significant changes upon cooling the sample to 100 K. It appears thus that the magnetic ordering in the films does not vary between 100 K and 300 K, suggesting that the ordering temperature of the trilayers is above room temperature. An ordering temperature significantly higher than in the bulk has also been found for thin Cr films in Cr/Fe multilayers[19,20] and it is attributed to proximity effects.[20,21]

The fcc Co/Fe/Co(100) trilayers present indeed a few suggestive analogies to the bcc Fe/Cr/Fe(100) system. Notably, Fe acts as a nonferromagnetic spacer in the fcc Co/Fe/Co trilayers, whereas in the bcc Fe/Cr/Fe system it represents the ferromagnetic component. Both systems show a correlation between the oscillation of interlayer coupling and those of spacer magnetization. In both cases the coupling displays fast oscillatory structures[19] that appear to be related to the (antiferro)magnetic ordering of the spacer. In atomically smooth Cr films the fast oscillation period of 2 ML dominates the coupling up to 50 ML. The data presented here in Fig. 2(a) and Fig. 3(b) show a minimum-maximum separation of approximately 1 ML.

A coupling period, however, cannot be determined due to the limited thickness range of stability for fcc Fe films. Moreover, it is conceivable that the Fe magnetic structure depends in a complicated way on the film thickness and the coupling is therefore aperiodic. While a stack of bcc(100)-Cr planes is layerwise antiferromagnetic (possibly with an incommensurate spin wave), the fcc(100)-Fe stacked planes could tend to develop noncollinear ordering due to the geometrical frustration of the antiferromagnetic fcc structure. For a detailed understanding of the fcc Fe film systems, layer-resolved information on the moment size and orientation would be highly desirable. For a Co/7 ML Fe/Co(100) sandwich, layer-resolved measurements[22] give evidence for a noncollinear magnetic structure of the fcc Fe films. The close correspondence reported here between the magnetization of the Fe films on Co(100) and in Co/Fe/Co(100) trilayers, and the indirect coupling across the Fe films, suggests that noncollinear magnetism of fcc Fe might play a key role in the determining of all these properties.

IV. CONCLUSIONS

The magnetic structure of ultrathin fcc Fe/Co(100) and fcc Co/Fe/Co(100) wedge samples has been investigated with x-ray magnetic circular dichroism. The development of the Fe dichroism signals as a function of film thickness shows a remarkable similarity in ultrathin fcc Fe/Co(100) and fcc Co/Fe/Co(100) trilayers. Moreover, the fcc Fe films mediate an interlayer coupling between the Co layers, the sign of which closely reflects the thickness dependence of the fcc Fe film magnetization.

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