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Induced V and reduced Co magnetic moments at the V/Co interface

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Using x-ray magnetic circular dichroism (XMCD) the vanadium and cobalt magnetic moments at the V/Co interface have been extracted for different V and Co thicknesses. We find a large magnetic moment induced on the V that is coupled antiferromagnetic to the Co. For Co both the spin and orbital moments are reduced at the interface. The evolution of the magnetic moment together with the spectral changes in the V $L_{2,3}$ XMCD upon V deposition demonstrate the complexity of the V/Co interface as manifested in its physical properties. We show how the hybridization at the interface between the Co and V can lead to a transfer of the exchange interaction, resulting in the modified magnetic moments. The change in the number of Co $3d$ holes, extracted from x-ray absorption spectroscopy, shows that the charge transfer over the interface is small, indicating that it can not be responsible for the change in the magnetic moments.

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I. INTRODUCTION

The appearance of magnetic moments in elemental metals that are nonmagnetic in their bulk form provides a challenging subject for theoretical and experimental studies. The understanding and tailoring of the magnetic properties of new magnetic materials can have a direct technological impact, in particular for magneto-optical and high-density data storage devices. In this context, vanadium, which is paramagnetic in its bulk body-centered-cubic (bcc) form, is a very interesting element and subject of intense debate and extensive research. It has been predicted that vanadium could become magnetic through the modification of the electronic structure which can be achieved by changing the local geometry of V atoms in surfaces, clusters, and alloys, i.e. changing coordination number and neighbor distances.[1–5] On surfaces, vanadium has been predicted to be nonmagnetic,[1] ferromagnetic,[6] antiferromagnetic,[7–9] and paramagnetic,[10] whereas experimental results evidence ferromagnetic,[11] antiferromagnetic,[12,13] and nonmagnetic[14,15] behaviors. Also the results for the magnetic properties of small V clusters[1–4,12] are contradictory, and the magnetic properties of V compounds and alloys are attracting considerable attention. While the V/Fe system has been studied extensively,[16–23] less is known and understood about other systems,[24–29] in particular the V/Co system. In the last decade the oscillatory magnetic exchange coupling through V layers in V/Co multilayers has been studied. It was shown that, for the existence of antiferromagnetic coupling, the V period (the spacing between two Co layers) is 9 Å.[30] However, new studies reported contradictory results giving an interlayer ferromagnetic coupling independent of the V thickness.[31] A more recent work showed that the coupling could be strongly affected by the structure of the Co/V multilayers and by the sharpness of the V/Co interfaces.[32] This is not surprising since the interface is believed to play an important role in the oscillatory magnetic coupling. In the same way it was recently demonstrated that the transition temperature in V/Co superconducting multilayers oscillates with the Co thickness.[33] This shows that not only the V/Co interface might be of importance for phenomena such as the giant magnetoresistance (GMR), but also that the thickness of the Co and V layers might play an important role in superconductivity. Surprisingly, there is no study in the literature reported for the V/Co interface, despite its importance for the interesting phenomena mentioned above.

In this paper we present an x-ray absorption spectroscopy (XAS) and x-ray magnetic circular dichroism (XMCD) study of the V/Co interface. We show that the Co atoms induce a strong polarization of the V atoms. The V atoms are found to be coupled antiferromagnetically with the Co film, and the influence of the Co and V film thickness on the magnetic moments of both V and Co is investigated.

II. EXPERIMENT

The magnetic thin films were prepared and measured *in situ* and at room temperature. The Co evaporation was performed in an ultra-high vacuum (UHV) preparation chamber operating in the low-10^{-9} mbar while the V evaporation was done in the UHV analysis chamber which remained in the low-10^{-11} mbar during measurements. Controlled evaporation of V and Co was performed on a Cu(100) surface cleaned by standard cycles of Ar^+ sputtering and annealing. The Co evaporator was calibrated using a quartz microbalance and the thickness of the V films was determined by comparing the intensity jump at the L_3 edge in XAS with previous measurements.[25] We estimate the accuracy for the film thickness to be within 20%. Different types of samples with monolayer thicknesses were prepared, namely, nML V/mML Co/Cu with $n=0.15$, 0.45, and 0.9 and $m=4$ and 10. The chosen Co thicknesses correspond to a film for which the coercive field is not saturated (4-ML Co) and a film with maximum coercive field (10 ML).[34] Note that these Co thicknesses are far above the controversial low-coverage

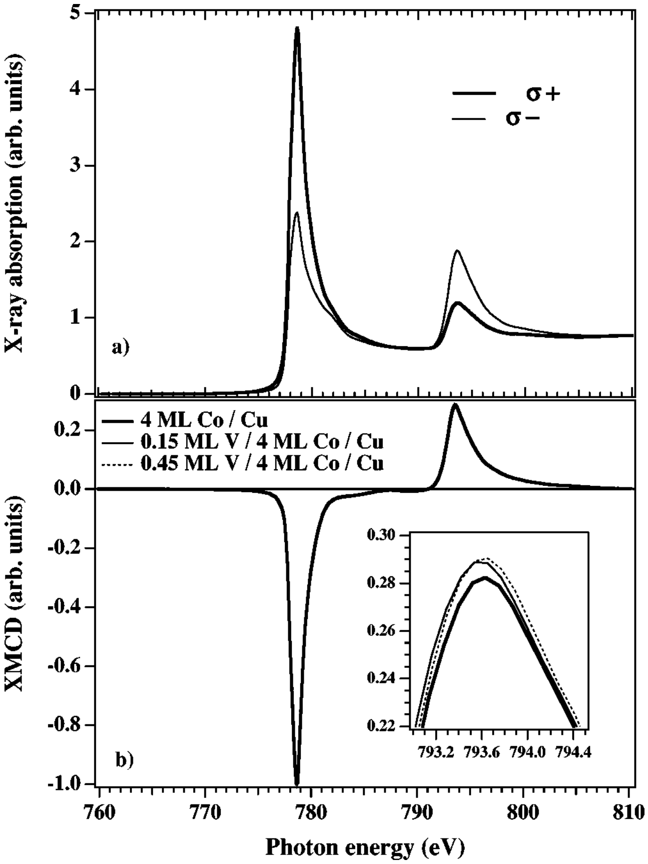

FIG. 1. (a) Typical Co $L_{2,3}$ absorption spectra for opposite magnetization directions. (b) Resulting difference spectra (XMCD); the displayed spectra have been normalized such that the XMCD at the Co L_3 peak is the same for all samples. The inset shows an enlargement of the XMCD at Co L_2 peak.

regime where metastable magnetic properties appear (<1.8ML),[35] and that the deposition temperature is below the threshold temperature for the segregation of Cu substrate atoms to the surface.[34]

XAS was measured in total electron yield detection mode on beamline ID08 of the European Synchrotron Radiation Facility (ESRF) at Grenoble, which provides monochromatized x-rays with a degree of circular polarization of $99.9 \pm 0.1\%$. A magnetic field of ± 7 T was applied parallel to the incident x-ray beam which was at normal incidence to the sample surface. The XMCD was obtained by taking the difference between the XAS with opposite magnetization directions. The absence of any contaminants (in particular of magnetic Cr, Fe, Ni, and Co) was carefully monitored using XAS.

III. RESULTS

A. Co $L_{2,3}$ XMCD and magnetic moments

Figure 1(a) displays the typical Co $L_{2,3}$ XAS measured for opposite magnetization directions. Figure 1b shows the normalized difference spectra, i.e., the XMCD, corresponding to different V coverages deposited on 4-ML Co. The figure displays for all samples the XMCD normalized to its minimum at the Co L_3 peak. As can be observed, the changes in the XMCD spectra are small and can be appreciated only in the inset showing the enhanced Co L_2 peak.

The Co magnetic moments were extracted from the XAS spectra with opposite magnetization using the sum rules.[36] The relation between the orbital moment, μ_L, spin moment, μ_S, magnetic dipole term, T_z, and number of $3d$ holes, n_h (all per atom), in the ground state and the intensities of the difference $\Delta A_{2,3}$ and sum $A_{2,3}$ spectra integrated over the corresponding $L_{2,3}$ edges is given by[36]

$$\frac{\mu_L}{n_h} = -\frac{4}{3}\,\frac{\Delta A_3 + \Delta A_2}{A_3 + A_2}, \tag{1}$$

$$\frac{\mu_S + 7T_z}{n_h} = -2\,\frac{\Delta A_3 - 2\Delta A_2}{A_3 + A_2}. \tag{2}$$

Theoretical results[37] indicate that $7T_z/\mu_S < 0.1$, hence giving an error in μ_S up to 10% by neglecting T_z. The extracted values for the Co magnetic moments per hole are shown in Table I. These values have been obtained assuming that $T_z = 0$ and that the mixing of the $2p_{3/2}$ and $2p_{1/2}$ states as well as the contribution of the sp valence band to the $L_{2,3}$ XMCD signal can be neglected.

TABLE I. Co orbital and spin magnetic moments, μ_L and μ_S, per $3d$ hole, their ratio, the number of $3d$ holes, n_h, and the Co and V total moments, $\mu_{\mathrm{tot}} = \mu_L + \mu_S$, per atom for different V/Co film thicknesses grown on Cu(100) as extracted from XMCD and XAS. All moments are given in μ_{B}. The V total moments were obtained assuming a constant number of $3d$ holes. The estimated error bars generated by the spectral analysis (background and double-step function subtraction, integration limits) are also indicated.

	Co μ_L/n_h	Co μ_S/n_h	Co μ_L/μ_S	Co n_h	Co μ_{tot}	V μ_{tot}
4-ML Co/Cu	0.121(1)	0.73(1)	0.165(4)	2.055(2)	1.74(3)	-
0.15-ML V/4-ML Co/Cu	0.106(1)	0.70(1)	0.151(4)	2.057(2)	1.65(3)	−1.8(1)
0.45-ML V/4-ML Co/Cu	0.090(1)	0.67(1)	0.134(3)	2.036(2)	1.54(3)	−1.5(1)
10-ML Co/Cu	0.117(1)	0.78(1)	0.150(3)	2.10	1.88(3)	-
0.15-ML V/10-ML Co/Cu	0.104(1)	0.79(1)	0.131(3)	2.118(1)	1.89(3)	−2.1(1)
0.45-ML V/10-ML Co/Cu	0.107(1)	0.77(1)	0.138(3)	2.109(1)	1.84(3)	−1.9(1)
7-ML Co/1.8-ML V/Cu	0.107(1)	0.69(1)	0.155(4)	2.932(2)	2.34(4)	−1.2(1)

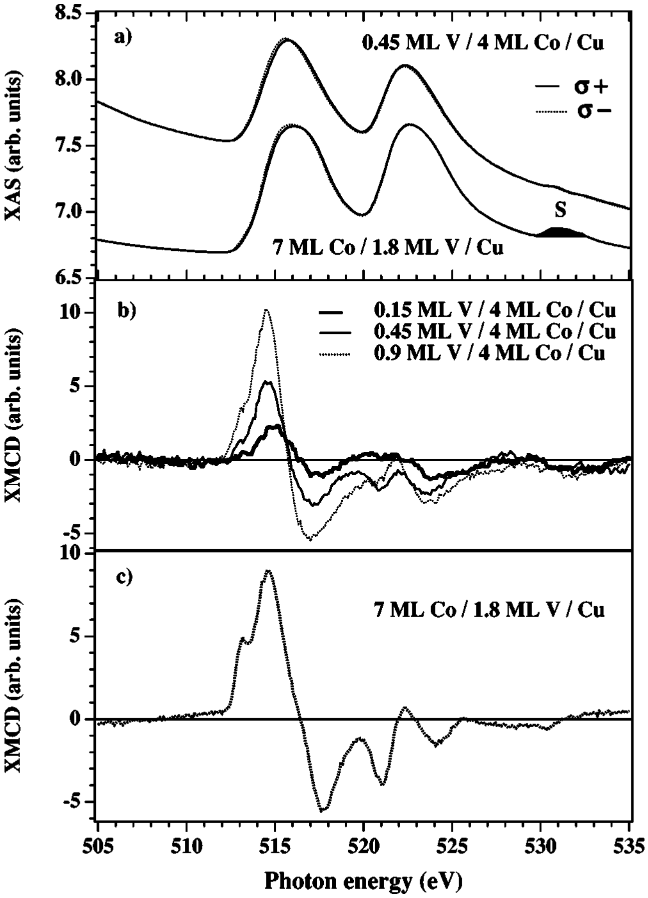

FIG. 2. (a) Typical V $L_{2,3}$ absorption spectra for opposite magnetization directions for both V/Co and Co/V interfaces. Note the presence of the satellite structure S in the case of the 7-ML Co/1.8-ML V interface. (b) Evolution of the resulting difference spectra (XMCD) with increasing V coverage. (c) XMCD corresponding to Co deposited on top of a V film.

B. Number of Co 3*d* holes

Since the XMCD sum rule analysis returns the values of μ_L/n_h and μ_S/n_h, we have to know the number of 3*d* holes to obtain the moments per atom. A knowledge of n_h is also needed to evaluate the influence of the charge transfer at the interface. We cannot assume that n_h would be the same at the interface as in the bulk, because the Co-V hybridization can induce a charge transfer between the two metals (cf. Sec. IV B). Therefore, n_h is expected to vary with layer thickness. According to the XAS sum rule[38] n_h is proportional to A_3+A_2. Following the procedure used in previous works[39,40] the XAS spectra were normalized to the intensity above the L_2 edge and the relative change in the integrated XAS signal was determined for the different systems. The 10-ML Co/Cu was taken as a reference, using the value of $n_h=2.1$ from the literature.[41] The values obtained for n_h of each system are given in Table I.

C. V $L_{2,3}$ XMCD

Figure 2(a) represents the typical V $L_{2,3}$ XAS of the V/Co measured for opposite magnetization directions. The XMCD signal in Fig. 2(b) clearly shows that there is a magnetic moment on the vanadium atoms. The sign of the V $L_{2,3}$ XMCD is opposite with respect to the Co $L_{2,3}$ XMCD in Fig. 1(b). Figure 2(b) shows that for increasing V coverage, the XMCD not only changes in size but also in spectral shape. This shape contains features of both the independent particle model[37,42] as well as the multiplet structure.[43] It is the combination of these two together that considerably complicates the explanation of the spectra. A further contribution to the shape might also arise from the relatively large magnetization of the *sp* band that is coupled parallel to the 3*d* magnetic moments in V. Although the study of the evolution of the V XMCD spectral shape goes beyond the scope of this paper, it is interesting to note that the XMCD spectrum for the lowest V coverage resembles those measured for V on Fe(100) (Ref. 15) and VFe alloy.[19] On the other hand, upon increasing coverage, the XMCD spectra show more structure and become more similar to the spectra measured for Fe/V multilayers[20,37] and superlattices.[21,22] This is especially the case for the V XMCD measured for the 7-ML Co on a thick V film (1.8 ML) where most of the features observed for the Fe/V multilayers[20,37] and superlattices[22] are reproduced.

D. Extracted V magnetic moment

For V the extraction of the magnetic moment from XMCD is more problematic than for Co. This is mainly due to the overlap of the L_3 and L_2 structures which are separated by a relatively small 2*p* spin-orbit splitting. Figure 2(a) shows that the XAS branching ratio, $L_3/(L_2+L_3)$, is close to 1/2. This is a rather large deviation from the statistical ratio of 2/3 which is due to the 2*p*-3*d* electrostatic interaction which is mixing the $j=3/2$ and 1/2 levels of the 2*p* core hole state.[44] Similarly, this *jj* mixing impedes the application of the second sum rule leading to large errors. To circumvent this, we extracted the magnetic moments following the procedure used for VCu alloys.[25] This procedure assumes that the maximum of the normalized dichroic signal is proportional to the magnetic moment with the scaling factor obtained using a reference sample with known magnetic moment. The extracted total moments are given in Table I. The above procedure might cause some systematic uncertainties and a detailed discussion on the validity and limitations of the procedure has been presented in Ref. 25. Note however that it has been used successfully for alloys with very low V concentration where μ_L and T_z can give large contributions[45] while here these contributions are expected to be smaller since the V concentrations are higher than for the $Cu_{96.7}V_{3.3}$ alloy.[25] As with the sum rule analysis [cf. Eqs. (1) and (2)] where the XMCD is normalized to the XAS, we are comparing the magnetic moments per hole rather than per atom. In order to obtain the total moments we have to make an assumption about the number of holes and we assumed that it is constant. The extracted n_h in Co for V/Co showed that the charge transfer is small. For V the relative change will even be smaller since there are more holes on the V. Therefore, this correction is not too important for the purpose of this study which concentrates on the trends in the V and Co magnetic moments per hole with film thickness.

IV. DISCUSSION

A. Magnetic moment trends

The magnetic moments obtained for the pure Co films (without V interface) are slightly higher than those reported for face-centered cubic (fcc) bulk Co.[46] However, as reported elsewhere, the Co spin and orbital magnetic moments for the Co/Cu(100) system show a non-trivial behavior depending on the thickness of the Co layer.[46–48] The discussion of this behavior is however beyond the scope of the present contribution.

As can be seen in Table I, upon V deposition the Co μ_S/n_h and μ_L/n_h are both decreasing and the ratio μ_L/μ_S is going down. This effect is more pronounced for the thinnest Co film, indicating that the effect is confined to the interface. On the other hand, the V moment decreases with increasing V coverage, as already reported for V/Fe(100).[15] This is probably due to the quenching of the V moments because of the increase in V neighbors and decrease of the average V-V distance.

The V moments are exceptionally high although overall they compare reasonably well with previous studies performed on V/Fe superlattices[21,22] ($\sim 1.5\mu_B$ and $\sim 1.1\mu_B$, respectively), hyperfine V particles[12] ($\sim 1.25\mu_B$) in the dilute V limit, in VCu alloys[25] and the calculated value for the VAu_4 alloy[49] ($\sim 1.7\mu_B$). The largest V atomic moment found here ($2.1\mu_B$) is far from those reported for the FeV system; however, it is not incompatible with the values predicted for free and supported V clusters.[3,50] Large V atomic magnetic moments are favored in our case since the V atoms are strongly polarized by the Co atoms while the V-V distance is increased due to the very low V coverage. It is interesting to note that like for V/Fe superlattices, the magnetic moment of V increases with decreasing V thickness and increases with increasing Fe (Co) thickness. Also remarkable is the strong reduction of the Co magnetic moment upon V deposition like for Fe magnetic moment in the V/Fe system.[21,22] These trends are illustrated in Fig. 3 showing the evolution of the V and Co atomic magnetic moments with the thickness of the Co and V films. Calculations for VFe indicate that the V orbital moment contributes only $\sim 2\%$ of the V total moment.[37] Assuming that this is also true for the VCo system, the strong reduction of the V moment upon V deposition of 10–15 % is likely to indicate that the V spin moment is drastically reduced upon increasing doses of V.

Without extracting the absolute moments, it is already possible to deduce from the overall spectral distribution of the V $L_{2,3}$ XMCD that the V spin and orbital moments are coupled antiparallel.[42] Comparing the signs of the XMCD shows us that V and Co have the orbital moments parallel and the spin moments antiparallel. Hence, the total moments of these metals are coupled antiparallel.

B. Influence of the hybridization

We shall now place the above quantitative results for the magnetic moments, as obtained from XMCD, in the “bigger picture” emphasizing the important role of the Co-V hybridization at the interface.

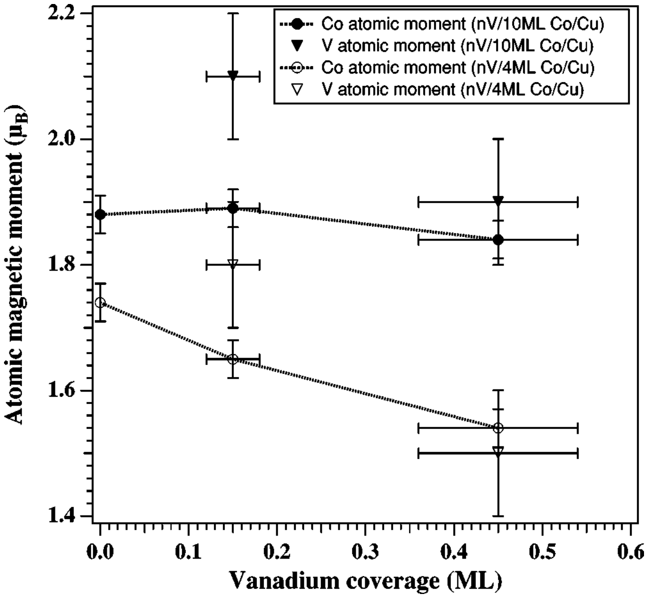

FIG. 3. Evolution of the V and Co atomic magnetic moments, μ_{tot}, with the thickness of the Co and V films as extracted from Table I. The V moments are shown with opposite sign.

1. Spin moments

The reduced spin moment for the Co, the induced spin moment for the V, as well as their antiparallel coupling, can all be understood as a direct consequence of the Co-V hybridization across the interface. In bulk Co, the $3d$ band is split into an almost filled spin-up (majority) band and a partly filled spin-down (minority) band. In bulk V, the partly filled $3d$ bands are degenerate in spin and located in energy above the Co bands. The centroids of the spin-up bands of Co and V are energetically well separated so that the hybridization (mixing) at the interface will be small. On the other hand, the centroids of spin-down bands of Co and V bands are energetically close together so that a relatively large hybridization between these bands develops at the interface. This is pushing down the Co spin-down band, thereby narrowing the distance to the Co spin-up band. In other words, the exchange splitting decreases, and the filling of the spin-down band will increase at the cost of the spin-up band, hence the Co spin moment will decrease. For V the hybridization results in a splitting of the spin bands with the spin-up band having a lower energy. Since the V $3d$ band is less than half filled this results in a V spin moment that is coupled antiparallel to the Co spin moment. It can be envisaged that this model of exchange-splitting transfer due to hybridization should apply generally at the interface of two metals. The effect is very large when one metal has a more than half-filled d band and the other a less than half-filled d band.[51] The experimental observation of a reduction in Co μ_S/n_h and the presence of an antiparallel spin moment on V upon interface formation is a clear proof of the hybridization effect. Furthermore, the decrease of the spin moments with V deposition clearly shows that the effect is restricted to the interface.

2. Orbital moment

Compared to the spin moment, the decrease in the Co orbital moment per hole at the interface is less straightforward to understand. The orbital moment depends strongly on the orbital symmetry of the spin-split states near the Fermi level within a typical energy range determined by the effective d spin-orbit interaction.[52] Similar to the exchange-splitting transfer (Sec. IV B 1), the hybridization will cause a transfer of the spin-orbit splitting at the interface between the two metals. For instance, it has been shown that at the Fe/W interface, where the W $5d$ spin-orbit interaction is much larger than that of the Fe $3d$, there is an enhancement of the Fe orbital moment.[51] Therefore, we can expect a reduction of the Co orbital moment at the Co/V interface, since the spin-orbit interaction of the V $3d$ is about three times smaller than that of the Co $3d$.

3. Charge transfer

Apart from a transfer of electrons from the spin-up to spin-down band in Co and a reversed transfer between the spin bands in V, we should also consider the possibility of a charge transfer between V and Co. Such a charge transfer could be expected from the difference in electronegativity between both metals that can lead to a transfer of electrons over the interface from V to Co. The excess nuclear charge displaces locally the mobile electrons close to the Fermi level, until the displaced charges screen out the nuclear charge exactly. This effect occurs at the interface over distances in the order of a few interatomic distances, i.e. the screening is strongly localized. The resulting change in the density of states near the Fermi level[39,53] can give a further modification of the spin and orbital magnetic moments in the magnetic layers. In Co with only few spin-up (majority) $3d$ electrons close to the Fermi level, the screening is almost uniquely due to spin-down (minority) $3d$ electrons. Relativistic local spin density calculations[51] typically give a value for the charge transfer of 0.1 electron for the first interface ML. This would correspond to a change in n_h of -4% for Co and $+2\%$ for V. In reality, these numbers could be much larger as was evidenced for the Ni/Co interface, where the experimentally observed change[53] in n_h largely exceeds the theoretically predicted value.[48]

The experimentally obtained number of Co $3d$ holes for the different V on Co systems extracted from XAS are shown in Table I. Compared to 10-ML Co/Cu, we find a slight decrease in n_h for 4-ML Co (-2%), 0.15-ML V/4-ML Co (-2%) and 0.45-ML V/4-ML Co (-3%), whereas no significant changes are observed for both 0.15- and 0.45-ML V on 10-ML Co. On the other hand, there is a strong increase for 7-ML Co/1.8-ML V (40%). These results indicate that the charge transfer from V to Co is very small when V is deposited on Co thin films but considerably larger in the opposite case, i.e., when Co is deposited on a relatively thick V film. This behavior could be due to structural effects of the V film (roughness, structural defects) that are known to modify significantly the electronic structure and magnetic properties.[32,48] Whereas it is well established that for coverages above 1 ML, V grows in four domains of bcc(110) on Cu(100)[54] and that Co grown on Cu(100) has a fct structure,[55–57] there is—to our knowledge—no structural or electronic study reporting on the properties of V/Co or Co/V interfaces. A forthcoming contribution[58] will address these specific issues.

Interestingly, we observed a strong satellite structure in the V $L_{2,3}$ absorption spectrum [labeled S in Fig. 2(a)] for the Co/V/Cu(100) interface that was not observed for the V/Co/Cu(100) interface. This satellite structure is located at a similar energy position as for the V/Fe system.[37] More investigation is needed in order to understand the origin of such a satellite structure, and in particular to correlate it to the presence (or absence) of a van Hove singularity at the N point of the bcc Brillouin zone, as recently suggested for the V/Fe system.[37] If the satellite is due to a van Hove singularity, its appearance could indicate a strong change in the electronic structure that can modify the charge transfer and, consequently, the magnetic moments.

V. CONCLUSIONS

Summarizing, we have presented an *in situ* study of the magnetic moments at the V/Co interface using XMCD. We found for submonolayer coverages that the Co induces a large magnetic moment on the V atoms. The evolution of the Co and V magnetic moments are in agreement with the expectations from a general model where the hybridization induces a transfer of the exchange splitting between the two metals. The reduced exchange splitting at the Co interface leads to an increase in the filling of spin-down (minority) states at the cost of the occupied spin-up (majority) states, hence reducing the Co spin moment per hole. The induced exchange splitting at the V results in an induced spin moment that is coupled antiparallel to the Co moment. The decrease in both the Co μ_S/n_h and V μ_{tot} with V deposition is a clear proof that this is an interface effect. The changes in the spectral shape of the V $L_{2,3}$ XMCD with Co and V thicknesses are highlighting the complexity of the Co/V interface. The V $L_{2,3}$ XMCD spectrum might also show a noticeable influence from the the spin polarized *sp* band, which would merit further study. Certainly, further experimental and theoretical work is required to obtain full understanding of these systems where we hope that the present study has contributed to the understanding of the Co/V interface and superlattices that are of interest for GMR and superconducting phenomena.

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