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Origin of Magnetic Contrast in Spin-Polarized Scanning Tunneling Spectroscopy: Experiments on Ultra-Thin Mn Films

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Normalized differential tunneling conductivities obtained with Fe-coated W tips show a spin-polarized peak around +0.8 V on ultrathin bct Mn films grown on Fe(001)-whiskers. This spin-polarized peak results in a clear magnetic contrast in spectroscopic images. Our normalization removes the influence of the tunneling probability and makes the spectroscopic curves most reliable for a derivation of the spin-resolved sample density of states (DOS) at positive voltages. From this analysis we conclude that the magnetic contrast in our spectroscopic maps is caused by a highly polarized DOS. Furthermore, a tip polarization of about 15% is found. [DOI: 10.1143/JJAP.42.4688]

KEYWORDS: iron, manganese, spin-polarized scanning tunneling microscopy, spin-polarized scanning tunneling spectroscopies, magnetic films, surface magnetism

1. Introduction

In the near future a lot of developments in the magnetic storage technology will be expected due to the present activation of magnetic and electronic studies in the nanometer range. Spin-polarized scanning tunneling microscopy (SP-STM) and spectroscopy (SP-STS) are the most powerful tools for studying surface magnetism with spatial resolution on the atomic scale.[1,2] Much progress has been made in SP-STM by Wiesendanger and coworkers.[1–6] One of the most important issues is to understand the origin of the magnetic contrast in STS images with magnetic tips. Although it was already reported that this magnetic contrast is related to the spin-polarized peak in spectroscopy,[5] this relation is not straightforward due to the influence of the tunneling probability. Kleiber *et al.* obtained the highest contrast in the dI/dV maps obtained on Cr(001) close to the surface state energy of -0.02 V but commented on a contrast inversion inherent to the closed-loop constant current mode which was used.[4] In a more recent publication, it was reported that the highest contrast is obtained around ± 0.25 V.[5] These effects are caused by the tip-sample separation dependence which is included in dI/dV. Recently, Okuno *et al.* used $(dI/dV)/(I/V)$ curves obtained on Co(0001) to extract the spin-polarization of the surface state.[7] However, it was shown by Ukraintsev that, although much better than dI/dV, this normalization cannot completely remove the tip-sample separation dependence.[8] Ukraintsev showed that, within a one-dimensional WKB approximation of tunneling, a normalization of dI/dV by its fitted tunneling probability function leads to the best recovery of the sample density of states (DOS) on the positive voltage side.

In this paper we show that a magnetic contrast can be obtained with Fe-coated W tips on ultrathin Mn films grown on Fe(001) whiskers, *whereas peaks are not observed in the* dI/dV *curves*. In addition we show that the normalization of dI/dV curves by tunneling probability functions does reveal a spin-polarized peak. We demonstrate experimentally that the spin-resolved surface DOS can be found using this normalization. With the help of band structure calculations we trace back the origin of the spin-polarized peak.

2. Experimental

STM and STS measurements were performed in ultrahigh vacuum ($\sim 5 \times 10^{-11}$ mbar) at room temperature (RT) with an Omicron UHV STM-1. Mn layers with a body-centered tetragonal (bct) structure were grown on an Fe(001)-whisker at 370 K. Details are given in refs. 9 and 10. In the present study, we used two kinds of tips: W tips (non-magnetic tips)[9] and Fe-coated W tips (magnetic tips).[11] A magnetic contrast was reproducibly obtained with W tips covered with 7–10 nm Fe at RT. Radii of our tips were more than 300 nm, which was measured by field emission spectroscopy. The magnetic contrast depends on the factor $\cos\theta$, where θ is an angle between the magnetization directions of the Fe-coated tip and the Mn layers. The tip magnetization direction is randomly orientated with respect to the magnetization direction of the Mn layers and is different from tip to tip. However, highest contrasts are observed when these magnetizations are parallel or antiparallel. Therefore, we believe that the largest experimentally observed magnetic contrasts (about 20 different Fe-coated tips were used) such as reported in this paper correspond to nearly (anti)parallel alignments. The tip was carefully brought close to the sample to exclude any mass transport between tip and sample. Furthermore, no voltage pulses were applied during these SP-STS measurements.[12] An external magnetic field was not applied. STS measurements were performed at every pixel of a constant current topographic image by opening the STM feedback loop at a given current and voltage ("setpoint"). dI/dV curves were obtained by numerical differentiation of the $I(V)$ curves. Band structure calculations were performed using the Vienna Ab initio Simulation Program (VASP).[13–15] An eight layer slab with the experimental values for the in-plane

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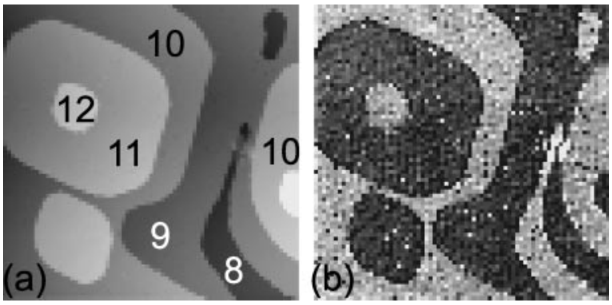

Fig. 1. SP-STM and SP-STS measurements on Fe(001) covered with about 10 ML Mn at 370 K. These measurements were performed with an Fe-coated W tip. (a) is the topographic image which was obtained at $V_S = -0.5$ V, $I = 0.5$ nA. Scan size is $100 \times 100\,\text{nm}^2$ and five layers are exposed. Numbers in Fig. 1(a) denote the stacking numbers of the Mn layers. (b) shows the dI/dV map at +0.2 V measured at the same area as (a), which was numerically obtained from dI/dV curves measured at a setpoint of $V_S = -0.5$ V, $I = 0.5$ nA.

(2.87 Å) and out-of-plane (2 × 1.64 Å) lattice constants was used. Furthermore, an antiferromagnetically stacking of the Mn(001) layers was assumed.

3. Results and Discussion

Since the growth of Mn layers on Fe(001) changes at coverages above 3 ML from layer-by-layer to layer-plus-islands,[9] five levels can be exposed at a coverage of about 7 ML Mn as shown in Fig. 1(a). Numbers in Fig. 1(a) denote the stacking numbers of the Mn layers. Previous results showed that from the fourth layer the Fe intermixing in the Mn film can be neglected.[9] At this surface, dI/dV curves were measured with Fe-coated W tips at 75 × 75 pixels at a setpoint of $V_S = -0.5$ V, $I = 0.5$ nA. At every pixel an $I(V)$ curve was measured which was numerically differentiated to obtain the dI/dV curve. The dI/dV map at +0.2 V in Fig. 1(b) shows a strong dark-bright contrast between the different levels. Clearly, the contrast oscillates with a period of two layers. This oscillating contrast starts from the fourth Mn adlayer (not shown here).[12] The contrast changes between dark (1.50 nA/V) and bright (1.85 nA/V) within a lateral length of 1.0 ± 0.5 nm.[16] This contrast was never observed with clean W tips:[9,11] dI/dV curves obtained on Mn layers with local coverages (Θ_L) larger than 3 monolayer (ML) are all equivalent.[16] In a recent publication this contrast was explained to be caused by spin-polarized tunneling into the antiferromagnetically coupled Mn(001) sheets.[11] Arguments for this interpretation are that the bright-dark contrast in the dI/dV maps is only observed with Fe-coated tips and that the onset ($\Theta_L > 3$ ML) and period (two layers) are in agreement with previous results obtained with other, non-local, techniques.[18–20] Furthermore, the strength and sign of the contrast (i.e., dark-bright might be reversed) depend strongly on the Fe-coated tip used which demonstrates the degree of freedom of the tip magnetization direction.

In the present paper we will analyze the origin of this contrast in more detail. Figure 2 shows dI/dV curves (solid curves) measured with an Fe-coated tip ($V_S = +0.5$ V, $I = 0.5$ nA) on Mn films with local coverages larger than 3 ML. dI/dV curves were obtained within a large voltage range from −2 V to +3 V. dI/dV curves obtained on the odd

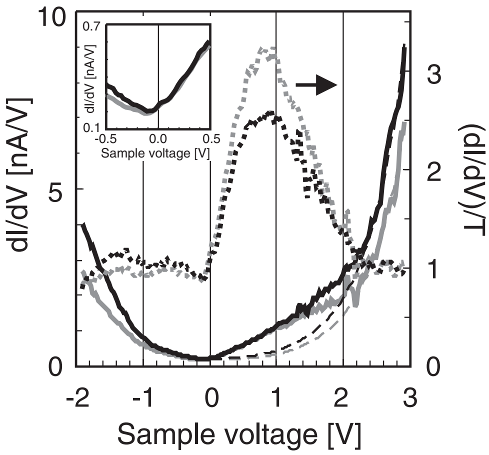

Fig. 2. dI/dV curves normalized by the voltage-dependent tunneling probability functions (T) were obtained with an Fe-coated W tip on Mn films thicker than three layers. Solid and dotted curves are the dI/dV and the $(dI/dV)/T$ curves, respectively. dI/dV curves averaged over 30 single curves were obtained at a setpoint of $V_S = +0.5$ V, $I = 0.5$ nA. T (dashed curves) were obtained by a fit to the dI/dV curves. Black and grey curves are representative of the odd and the even layers, respectively. An inset in the top-left of this figure shows dI/dV curves of odd and even layers within a voltage range from −0.5 V to +0.5 V. No difference is observed around +0.2 V.

(black) and the even (grey) layers show as a function of bias voltage a steady increase above the Fermi level and an exponential increase above +2 V. These dI/dV curves do not show any peak. Spectroscopic measurements in Figs. 1 and 2 were performed at voltage-setpoints of $V_S = -0.5$ V and $V_S = +0.5$ V ($I = 0.5$ nA), respectively. Different setpoints cause different tip-sample separations, which has a strong influence on the dI/dV signals. This is the reason why the dI/dV curves of Fig. 2 are the same at +0.2 V, whereas the dI/dV map of Fig. 1(b) shows a clear contrast at +0.2 V. To remove the tip-sample separation dependence, dI/dV curves were normalized by tunneling probability functions (T). Theoretically, this normalization was shown to lead to the best sample DOS recovery.[8] Also, our recent publications show that this method can successfully recover the sample DOS from experimental dI/dV curves.[9,10] The tip-sample separation is included in the tunneling probability function:

$$T = K \exp\left[-2S\left(\frac{2m}{\hbar^2}(\bar{\Phi} - eV/2)\right)^{1/2}\right], \qquad (1)$$

where $V > 0$. K is a proportionality coefficient related to the effective tip-sample contact area. $\bar{\Phi}$ is the average of sample and tip barrier height, S the tip-sample separation, e the electron charge, and m the electron mass. We fitted T multiplied by an offsetted Gaussian line shape, representing the surface state peak in the DOS, to the dI/dV curves. Tunneling probability functions for odd (T_{odd}) and even (T_{even}) layers are shown as dashed curves in Fig. 2. The normalized dI/dV curves ($(dI/dV)/T$: dotted curves in Fig. 2) show a strong peak around +0.8 V. The peak amplitude oscillates with a period of two layers. Since the Mn layers couple antiferromagnetically and only magnetic tips show these characteristics, we can conclude that this oscillating amplitude of the peak is caused by spin-polarized tunneling.

An interesting point is the origin of this spin-dependent peak at +0.8 V. To elucidate this we will use the approximation made in ref. 8 to recover the spin-resolved DOS from our normalized dI/dV curves. For positive sample bias voltages, the dI/dV curves for the odd and the even layers can be described as follows:

$$\begin{aligned}(dI/dV)_{\rm even} &= (D^{\uparrow}_{\rm Fe}\cdot D^{\rm min}_{\rm Mn} + D^{\downarrow}_{\rm Fe}\cdot D^{\rm maj}_{\rm Mn})\cdot T_{\rm even}\\ (dI/dV)_{\rm odd} &= (D^{\uparrow}_{\rm Fe}\cdot D^{\rm maj}_{\rm Mn} + D^{\downarrow}_{\rm Fe}\cdot D^{\rm min}_{\rm Mn})\cdot T_{\rm odd}.\end{aligned} \quad (2)$$

$D^{\uparrow}_{\rm Fe}$ ($D^{\downarrow}_{\rm Fe}$) indicates the DOS at the Fermi level (E_F) for the majority (minority) bands of the ferromagnetic Fe-coated tip. Here, since Fe is ferromagnetic, majority-spin and minority-spin are defined as spin-up and spin-down. $D^{\rm maj}_{\rm Mn}$ ($D^{\rm min}_{\rm Mn}$) indicates the DOS at ($E_F + eV$) for the majority (minority) bands of the sublattices of antiferromagnetic bct Mn films. Since spin-up and spin-down are not defined for antiferromagnetic slabs, we prefer to use majority and minority DOS for the Mn layers, instead. In eq. (2) we assume that spin-up(-down) electrons of the Fe-coated tip tunnel into the minority-(majority-)spin bands of the odd Mn layers and the majority-(minority-)spin bands of the even Mn layers without spin-flipping. However, remember that the tip magnetization direction is random and might be either parallel to the even or odd Mn layers. Using the even and the odd $(dI/dV)/T$ curves in Fig. 2, the averaged $(dI/dV)/T$ curve and the asymmetry in $(dI/dV)/T$ are obtained (Fig. 3(a)). The asymmetry ($A_{(dI/dV)/T}$) is defined as follows:

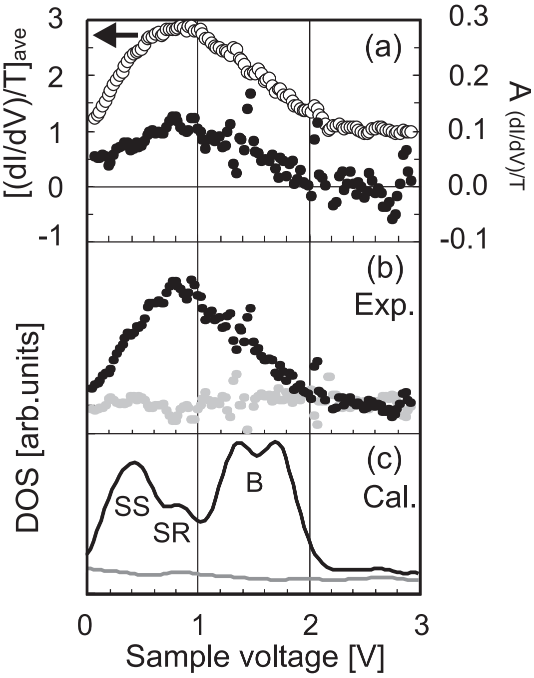

Fig. 3. (a) shows the averaged dI/dV curve of the odd and the even $(dI/dV)/T$ curves in Fig. 2 (white circles) and the asymmetry of the $(dI/dV)/T$ curves obtained by eq. (3) (black dots). (b) shows the majority (grey) and minority (black) DOS of Mn(001) obtained by experimentally obtained $(dI/dV)/T$ curves (Fig. 2). (c) shows the majority (grey) and minority (black) DOS of Mn(001) obtained by band structure calculations.

$$\begin{aligned}A_{(dI/dV)/T} &= \frac{(dI/dV)_{\rm even}/T_{\rm even} - (dI/dV)_{\rm odd}/T_{\rm odd}}{(dI/dV)_{\rm even}/T_{\rm even} + (dI/dV)_{\rm odd}/T_{\rm odd}}\\ &= \frac{D^{\uparrow}_{\rm Fe} - D^{\downarrow}_{\rm Fe}}{D^{\uparrow}_{\rm Fe} + D^{\downarrow}_{\rm Fe}}\cdot\frac{D^{\rm min}_{\rm Mn} - D^{\rm maj}_{\rm Mn}}{D^{\rm min}_{\rm Mn} + D^{\rm maj}_{\rm Mn}}\\ &= P_T(E_F)\cdot P_S(E_F + eV), \quad \text{where} \quad V > 0.\end{aligned} \quad (3)$$

$(dI/dV)_{\rm even}/T_{\rm even}$ and $(dI/dV)_{\rm odd}/T_{\rm odd}$ are $(dI/dV)/T$ curves obtained on the even and the odd layers, respectively, P_T the tip polarization at E_F, P_S the sample polarization at ($E_F + eV$). $A_{(dI/dV)/T}$ shows a maximum value of about 10% around the peak energy and reaches 0% above +2 V (Fig. 3(a)). Using eqs. (2) and (3) the spin-resolved DOS of Mn(001) can be experimentally obtained as follows:

$$\begin{aligned}D^{\rm maj}_{\rm Mn} &= \frac{1}{c}\left(1 - \frac{A_{(dI/dV)/T}}{P_T}\right)[(dI/dV)/T]_{\rm ave}\\ D^{\rm min}_{\rm Mn} &= \frac{1}{c}\left(1 + \frac{A_{(dI/dV)/T}}{P_T}\right)[(dI/dV)/T]_{\rm ave},\end{aligned} \quad (4)$$

where $[(dI/dV)/T]_{\rm ave}$ is the average of $(dI/dV)_{\rm odd}/T_{\rm odd}$ and $(dI/dV)_{\rm even}/T_{\rm even}$. c is the sum of $D^{\uparrow}_{\rm Fe}$ and $D^{\downarrow}_{\rm Fe}$.

By following eq. (4), the majority (grey) and the minority (black) DOS of Mn(001) can be obtained experimentally as shown in Fig. 3(b). A broad peak is observed around +0.8 V only in the minority DOS. Figure 3(c) shows the majority and the minority local DOS at the surface of Mn(001) obtained by band structure calculations.[11] There are peaks in the minority DOS, while the majority DOS is low and flat without any structure above the Fermi energy. In Fig. 3(c), "SS", "SR", and "B" refer to surface states, surface resonance, and bulk states, respectively. Fitting of the experimentally observed majority DOS to the calculated DOS leads to a tip polarization of 15%. Qualitatively, Fig. 3(b) is in agreement with Fig. 3(c). This gives evidence that the spin-dependent peak at +0.8 V in the $(dI/dV)/T$ curves is caused by this highly polarized DOS. Although, from Fig. 3(b), a tip polarization of 15% is found, the polarization of an Fe-coated tip was assumed to be as high as 44% in ref. 6. When we use such a high tip polarization, our experimental results produce a pronounced peak in the majority DOS which is qualitatively inconsistent with our theoretical calculation. Also, this high tip polarization (~40%) likely includes uncertainties; (1) This high tip polarization was concluded from tunneling measurements of Fe/Al_2O_3/super-conductor junctions,[21] but the barrier materials greatly influence the measured spin polarization.[22] (2)The spin polarization of Cr(001) was deduced to be 17% by assuming about 40% polarization for the Fe-coated tip[4] whereas Cr(001) has highly spin-polarized *d* surface states.[23]

4. Conclusion

A magnetic contrast was obtained on ultrathin bct Mn films grown on Fe(001) whiskers by SP-STM with Fe-coated W tips. Normalizing the dI/dV curves by the tunneling probability function removed the tip-sample separation dependence and revealed a spin-polarized peak at +0.8 V. We demonstrated that the spin-resolved sample DOS which we extracted from the $(dI/dV)/T$ curves is in agreement with our band structure calculations. Consequently, the magnetic contrast in the SP-STS can be explained by this highly SP-DOS around +0.8 eV. A tip polarization of around 15% can be derived by a comparison between the calculated DOS and experimentally obtained DOS.

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