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Spin-polarized scanning tunneling microscopy of the room-temperature antiferromagnet *c*-FeSi

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Antiferromagnetic spin ordering has been revealed by room-temperature spin-polarized scanning tunneling microscopy (SP-STM) in thin epitaxial films of *c*-FeSi on Si(111). Spin polarization of tunneling current for unoccupied states is found to be unusually large $I_{\uparrow\uparrow}/I_{\downarrow\uparrow} = 3.8$. Atomically sharp spin-frustration domain walls, developing on the surfaces of nanoscale islands, have been observed on SP-STM images. Our results suggest that antiferromagnetism in *c*-FeSi is driven by Mott-Hubbard transition, and the atomically narrow domain walls are caused by local insulator-to-metal breakdown.

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Design of thin film magnetic materials capable of storing information at the atomic level represents one of the far-reaching goals of nanotechnology. The experimental study of atomic scale magnetism can be performed using spin-polarized scanning tunneling microscopy (SP-STM),[1–8] the technique combining the tunneling magnetoresistance (TMR) effect[9] with high lateral resolution of STM. So far, SP-STM has been credited for the study of domain walls in ferromagnetic nanostructures,[1,2] spin order on antiferromagnetic surfaces,[3] spin frustration induced by topological defects,[4–6] spin polarization in single-atom contacts,[7] and bistability in artificial atomic scale antiferromagnets created by the atom manipulation technique.[8] The most recent of these SP-STM works[8] has shown that magnetic information can be stored on surfaces using 12-atom magnetic bits.

In this Rapid Communication we will show that even more dense storage of magnetic information can be achieved using antiferromagnetic thin films. We will describe our SP-STM study of *c*-FeSi thin epitaxial islands on the surface of Si(111). In our study, we (a) revealed the existence of antiferromagnetic spin order in this material; (b) obtained atomically resolved structural and magnetic images and observed atomically narrow spin-domain walls on surface of this material indicating its potential application for atomic scale magnetic memory storage; (c) found unusually high for SP-STM spin polarization of tunneling current; and (d) observed spectroscopic signatures of Hubbard bands. We show that antiferromagnetism in *c*-FeSi is most likely driven by Mott-Hubbard transition, and atomically narrow domain walls arise from local insulator-to-metal breakdown.

The experiments were performed in an ultrahigh vacuum (UHV) system with a base pressure of 1×10^{-10} Torr equipped with STM, metal deposition sources, ion gun, and surface analysis tools. The surface of the Si(111) substrate was cleaned by sequence of UHV annealing to 1200 °C and ion beam sputtering. The quality of the surface was monitored by reflective high energy electron diffraction (RHEED), Auger spectroscopy, and STM. After cleaning, a 1-nm-thick Fe film was deposited *in situ* on this substrate at room temperature from an electron-beam evaporator. After deposition, the substrate was annealed to 500 °C for 30 s to form a thin layer of iron-silicide [see Fig. 1(c)]. This process, as shown in earlier published STM studies,[10–14] creates the islands of epitaxial silicide, whose top surfaces are atomically flat. For growth conditions reported here, the epitaxial growth of (111)-oriented flat-top nanoscale islands of *c*-FeSi was documented in the literature.[10–14] This material, known to exist only as epitaxially stabilized thin film, has a cubic unit cell with CsCl-type structure and $a = 1.5$ Å spacing between adjacent (111)-oriented atomic layers. In STM experiments the hexagonal packing of atoms on (2×2) reconstructed surface allows us to identify these (111)-oriented islands. The interest in magnetic properties of *c*-FeSi arose after the discovery of anomalous antiferromagnetic (AFM) coupling in Fe/*c*-FeSi/Fe multilayers[15–21] resembling magnetic behavior of Fe/Cr/Fe,[22] which indirectly suggested the possibility of room-temperature antiferromagnetic spin ordering in this material: A spin ordering never directly observed so far. For spin-polarized STM measurements, we used an electrochemically etched tungsten tip, whose surface was cleaned in UHV using the field emission technique, and whose spin-polarized apex was *in situ* fabricated using field-induced evaporation. For this purpose, a sequence of negative voltage pulses was applied on a STM tip. Fe atoms from the terminating atomic layer, which were transferred by this process, formed an atomically sharp high-aspect-ratio tip apex. During STM measurements, the sample was kept at room temperature.

In Fig. 1(a) we show the 180×180 Å^2 STM image obtained on top of a *c*-FeSi island. During this measurement, the tip voltage was kept at +0.5 V. In the central portion of the image we observe a vertical nanopipe aligned with a core axis of a growth spiral. The period of a spiral terrace, as determined from the cross section in Fig. 1(e), is $d = 4.5$ Å that corresponds to three atomic layers of *c*-FeSi. The hexagonal packing of atoms, which can also be observed in Fig. 1(a), clearly indicates the (111)-oriented growth of the *c*-FeSi epitaxial film. The lateral atomic corrugation has a periodicity of 7.2 Å as is indeed anticipated for the (2×2) reconstructed (111) surface of *c*-FeSi. Upon sign reversal of the tip voltage, instead of imaging occupied states in the sample, as in Fig. 1(a), the imaging of empty states takes place. The corresponding STM image of *c*-FeSi is presented in Fig. 1(b) that was obtained at the tip bias of −0.75 V. In addition to structural features, earlier seen in Fig. 1(a), here we observe an atomically sharp spin domain boundary originating at the axis of a growth spiral. A portion of surface [area 1 in

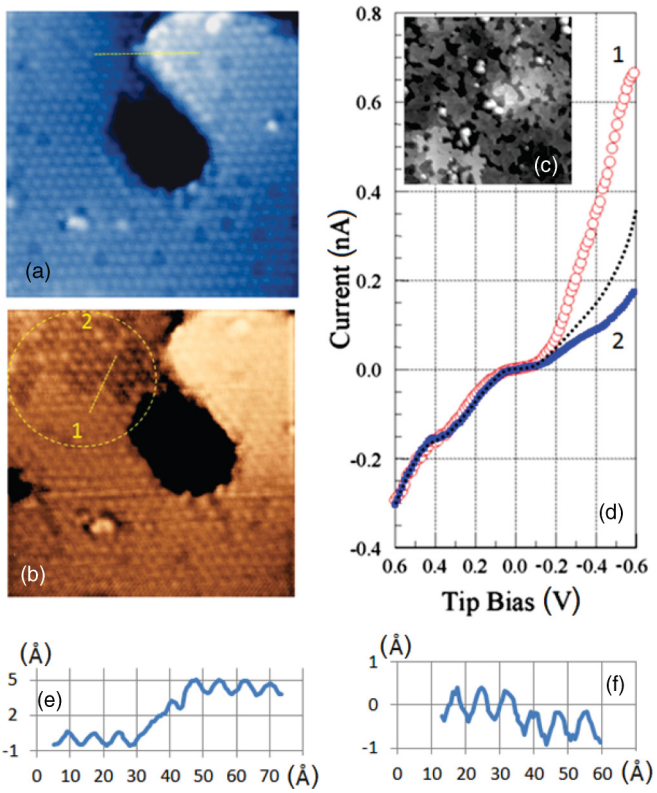

FIG. 1. (Color online) (a) and (b) The 180 × 180 Å^2 STM images of (a) occupied states at $V_{tip} = +0.5$ V and (b) empty states at $V_{tip} = -0.75$ V in c-FeSi thin epitaxial island on Si(111) surface. (c) Large scale, 0.7 × 0.7 μm^2, STM image of studied sample. (d) Curves 1 and 2: Tunneling I-V characteristics recorded with spin-polarized STM tip in areas 1 and 2 [see (b)] at the opposite sides of a spin boundary. Dotted line: Tunneling I-V characteristic recorded before making a spin-polarized STM tip. (e) The horizontal cross section of a spiral terrace in (a). (f) The cross section of a spin boundary in (b).

Fig. 1(b)] whose magnetization coincides with magnetization of the STM tip is $\delta z = 0.5$ Å higher than the other portion of surface [area 2 in Fig. 1(b)], whose magnetization is opposite to magnetization of a probe. This purely electronic (nonstructural) step in the STM image can also be seen in the cross section shown in Fig. 1(f). Surprisingly, the zigzag-shaped spin-reversal boundary is found to be both atomically sharp and spatially pinned to the lattice of the surface atoms. According to earlier published SP-STM works,[23,24] the bias dependence of spin-polarized STM images develops due to energy dependent overlapping of spin polarizations for the STM tip and sample.

Our experiments show that on top of c-FeSi islands surface atoms and surface spins can be visualized simultaneously using a constant-current topographic mode of STM. The height contrast $\delta z = 0.5$ Å, which develops at a boundary of oppositely polarized domains, originates from the difference of tunnel current-voltage characteristics in surface areas 1 and 2. These current-voltage characteristics are shown in Fig. 1(d). For positive tip voltages, corresponding to occupied states in a sample, the tunneling I-V characteristics essentially coincide. Imaging c-FeSi islands in this range of voltages brings only structural information about the arrangement of surface atoms. At negative tip voltages, corresponding to unoccupied states in a c-FeSi sample, we observe a significant difference in tunneling I-V curves for oppositely polarized domains. Now the ratio of tunneling currents corresponding to parallel and antiparallel spin orientations is $I_{\uparrow\uparrow}/I_{\downarrow\uparrow} = 3.8$, which is significantly higher than usually reported in SP-STM literature.[1–8] As we shall show later, the unusually strong spin polarization of tunneling current has to do with strongly correlated AFM Mott-Hubbard ground state in c-FeSi. Due to the exponential dependence of tunneling current on tip-to-sample distance $I \sim \exp(-\alpha z)$ on STM images of empty states the magnetic boundary must be seen as a sharp vertical step, whose height $\delta z = \alpha^{-1} \ln(3.8) \approx 0.6$ Å, assuming that $\alpha \approx 2$ Å^{-1} as it is typical for UHV tunneling. This unusually strong spin polarization of tunneling current makes it possible to directly observe a superposition of atomically resolved structural and magnetic images using constant-current STM mode in our experiment. From the experimentally determined value of $\gamma = I_{\uparrow\uparrow}/I_{\downarrow\uparrow}$ using Julliere's model[9] we can estimate the product of spin polarizations of electronic density of states (DOS) in the studied sample (P_S) and in the STM tip (P_T) according to

$$P_S P_T = \frac{\gamma - 1}{\gamma + 1} \tag{1}$$

that yields $P_S P_T = 0.58$.

For reference, in Fig. 1(d) we also show the tunneling I-V characteristic obtained on the surface of the island *before* making a spin-polarized tip apex [dotted line in Fig. 1(d)]. This curve is more symmetric than spin-polarized curves, and it essentially can be viewed as a "spin average" of tunneling I-V curves 1 and 2 in the same Fig. 1(d). The asymmetry of spin-polarized I-V characteristics arises from enhancement or suppression of tunneling current at a negative tip bias due to TMR effect. The spin-polarized I-V characteristics (1 and 2), shown in Fig. 1(d), have been reproducibly observed on different c-FeSi islands. Usually only one type of I-V curve can be measured on an atomically flat island: Either type 1 or type 2 depending on spin polarization of the topmost atomic layer.[25]

In Fig. 2(a) we show the elementary unit cell of c-FeSi constructed under the assumption of antiparallel spin alignment for nearest (2.6 Å spaced) Fe atoms.[26] Our construction clearly shows the development of constant-polarization (111)-oriented atomic planes, with out-of-plane (i.e., [111]) magnetic periodicity of $l = 2a = 3.0$ Å. In a spiral terrace geometry, shown in Fig. 2(b), spin frustration occurs if the vertical period of a spiral structure (d) equals half-integer number of magnetic periods (l),

$$d = (n + 1/2)l. \tag{2}$$

Please note that the drawing in Fig. 2(b) corresponds to a simplest case of $n = 0$, whereas for a real island in Fig. 1(a) $d = 4.5$ Å and $n = 1$. As a consequence of topology-induced spin frustration, the top surface of the island reveals atomically sharp spin-reversal domain boundary, which has been observed in our experiment using SP-STM. The spin-reversal boundary for c-FeSi is found to be significantly narrower than 6–8 atomic rows earlier reported for antiferromagnetic Fe monolayer on

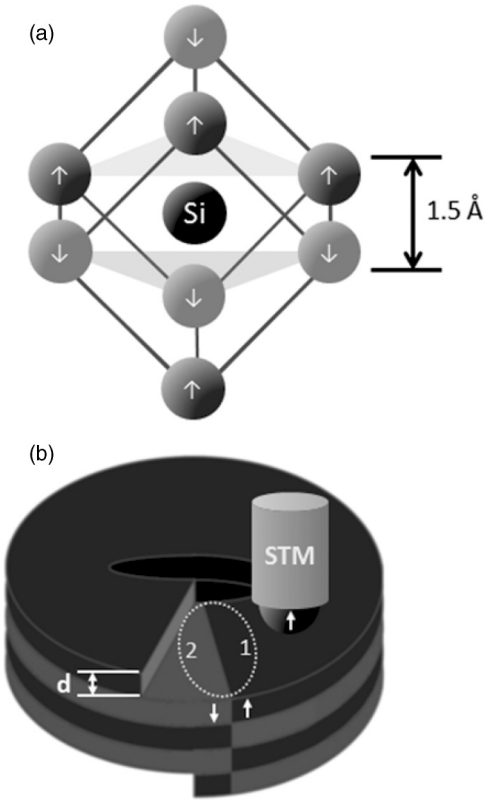

FIG. 2. (a) The elementary unit cell of *c*-FeSi, with CsCl-type structure, constructed under the assumption of AFM coupling between nearest Fe atoms. Spin polarization of Fe atoms is indicated by arrows. The [111]-oriented magnetic period equals 3.0 Å. (b) The topology-induced spin frustration in a spiral terrace geometry for a simplest case of $n = 0$. The areas 1 and 2 on this figure correspond to similar numbered areas on the STM image in Fig. 1(b).

W(001),[3] but it is comparable to an earlier reported wall width for Fe nanostripes.[1,27] It is well known that the width of a magnetic domain wall essentially represents the smallest magnetic domain size.[28] The combination of magnetic properties of *c*-FeSi, found in our experiments, including (a) atomically small domain size, (b) relatively high spin-ordering temperature, and (c) high degree of spin polarization, suggests this material as a good practical candidate for atomic-scale magnetic memory storage. For example, in Fig. 1(b) one can see small metastable spin domains (in area 2) nucleated during the growth of the film with lateral sizes of 2×2-reconstructed surface unit cell and comprised of four atoms. We cannot comment here why AFM spin ordering in *c*-FeSi was not observed in earlier works.[15–21] Perhaps, because SP-STM is a surface sensitive technique for *in situ* study of materials grown in ultraclean UHV environment, and is capable of spatially mapping their topography and spin polarization at atomic scale, it can accomplish research tasks that other methods cannot.[29]

With respect to the nature of AFM ground state found in *c*-FeSi, we believe this material represents a Mott-Hubbard type antiferromagnet similar to NiO.[30] This is also consistent with our observation of a nearly symmetric pair of steplike features in the tunneling I-V spectra of *c*-FeSi [see, for example, curve 2 in Fig. 1(d)] at -0.3 and $+0.3$ V. The corresponding DOS

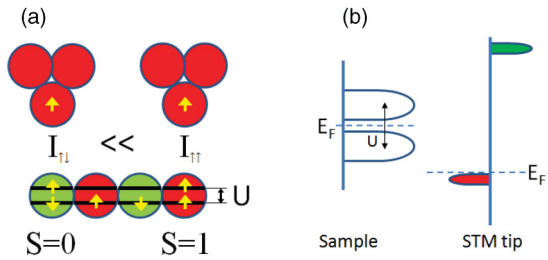

FIG. 3. (Color online) (a) Tunneling from metallic SP-STM tip into unoccupied Hubbard band leads to formation of excited state with the total spin $S = 1$ or $S = 0$ depending on relative spin alignment for the STM tip and sample. (b) Schematic band diagram of our experiment. Only spin-polarized bands are shown for the STM tip. The spin-majority band is located just below the Fermi level of the STM tip, while the spin-minority band is located well above the Fermi level. The splitting of magnetic bands is consistent with the data for Fe films from Ref. 33.

maxima are likely to be related to occupied and unoccupied Hubbard bands[31] in *c*-FeSi, with Hubbard $U \approx 0.5$ eV. This observation indicates that the electronic structure of *c*-FeSi has been affected by Mott-Hubbard transition: A transition that leads to AFM spin ordering. Because SP-STM studies of AFM Mott-Hubbard insulators were not conducted so far, in the space below we would like to clarify how the STM spin sensitivity develops for such materials on the atomic scale. Our understanding of this mechanism is based on a widely used assumption[32] that tunneling into strongly correlated electron systems represents the process of electron *addition* or *removal* accompanied by formation of excited atomlike states. In Fig. 3(b) we show the schematic band diagram of our experiment. Spin-polarized electrons tunneling from the STM tip are supplied by narrow magnetic band located just below the Fermi level. As we show in Fig. 3(a), tunneling into unoccupied Hubbard band is accompanied by formation of excited state with total spin $S = 1$ or $S = 0$ depending on relative spin alignment for the STM tip and sample. Because only $S = 1$ state is allowed by Hund's rule, tunneling current for this geometry enhances, while for opposite geometry it decreases. As a result, for negative polarity of tip bias we have $I_{\uparrow\uparrow}/I_{\uparrow\downarrow} \gg 1$. This ratio is limited only by spin polarization of STM tip $P_T = 0.58$. For opposite tip bias polarity, electrons tunnel from the occupied Hubbard band into the empty states in the STM tip: The states that are not spin polarized, as we show in Fig. 3(b). As a result, we have $I_{\uparrow\uparrow}/I_{\uparrow\downarrow} = 1$, in agreement with our experimental data. With respect to the atomically narrow domain walls observed in Fig. 1(b), usually the Bloch wall width is determined by

$$\delta_B \sim \sqrt{A/K}, \tag{3}$$

where A and K are exchange and magnetic anisotropy constants,[28] and $\delta_B \sim$10–100 nm. For Mott-Hubbard AFM, atomically sharp ($\delta \ll \delta_B$) antiphase domain boundaries must be considered as "electronic defects," that is, regions where metal-insulator transition is locally suppressed. Because the activation energy of such electronic defects must be less than the Bloch wall energy ($\sim\sqrt{AK}$), atomically narrow domain walls are likely to develop when the Mott-Hubbard localization

parameter[32] t/U is not far from its critical value. Under such conditions, the domain wall is filled with dense Femi gas, which totally screens the interwall exchange coupling. Thus, our approach to the physics of atomically sharp spin domain walls basically relies on the concept of spatial phase separation in Mott-Hubbard insulators.[34,35]

In conclusion, our SP-STM experiments revealed previously unknown antiferromagnetic spin order in c-FeSi accompanied by (a) unusually large tunneling spin contrast and (b) atomically narrow domain boundaries. The data analysis suggests that the observed spin order has to do with strongly correlated electronic state arising from metal-insulator transition, most likely Mott-Hubbard transition.

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