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A high-reflectivity atom-focusing mirror stable at room temperature

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It is shown that the Pb($\sqrt{3}\times\sqrt{3}$)R30/Si(111) ordered layer is an excellent mirror for neutral He atoms. It focuses more than 15% of the incoming He atoms into the specular peak, and is stable up to 450 K. Moreover, the reflectivity remains almost unchanged in a time scale of several weeks in ultrahigh vacuum. As a consequence, this system is a very good candidate to be used as a mirror in the next generation of the scanning helium atom microscope.
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There is no doubt that the invention of imaging techniques such as scanning tunneling microscopy (STM) brought a revolution in surface science, which led in particular to the birth of the field of nanotechnology. However, in several branches of nanotechnology there is still a need for a technique able for imaging insulating glass surfaces, delicate biological materials, and fragile samples which are difficult to examine by STM due to sample charging or electron excitation effects.

Perhaps the most important of these techniques, currently under development, is the scanning helium atom microscope,[1,2] which uses a focused beam of neutral He atoms as imaging probe. Such a microscope would be a unique tool for reflection or transmission microscopy, since—due to the low energies used ($\sim$100 meV)—neutral He atoms probe the topmost surface layer of any material in a completely nondestructive manner.[3,4]

The practical realization of such a microscope requires the development of a mirror able to focus a beam of low energy He atoms into a small spot on the sample to be examined. Holst and Allison[1] demonstrated that electrostatic bending of a thin, H-passivated Si(111)-(1×1) crystal was able to focus a 2 mm-He beam to a spot diameter of 210 microns. In practice, an ellipsoidal mirror is created by electrostatic bending of a single crystal silicon wafer. To provide the necessary elastic properties for repeated bending, the single crystal substrate must be very thin, about 50 μm thick, which cannot be achieved with metallic crystals.[5,6] A serious limitation, however, to improve the resolution was the low intensity obtained in the focused peak, which is a consequence of the poor reflectivity of such surfaces, less than 1%.

Whereas the potential lateral resolution of this microscope is $\sim$50 nm, the best value reported using current prototypes is 1.5 microns.[2,7] The main limitation to go beyond this value is given by the low reflectivity of the mirrors used to focus the incoming He atoms into the specular beam, which lies near 1% for H-passivated Si(111)-(1×1) mirrors. We have recently showed that reflectivities of ca. 15% can be obtained using Pb thin films stabilized by quantum size effects (QSEs) on Si(111) surfaces.[8–10] Whereas this result is very promising, a serious shortcoming is that such surfaces are stable only up to 250 K, introducing a practical limitation to mirror holders which should be kept below room temperature even when the microscope is not operating. In this letter we present further results which show that similar reflectivities can be obtained with an ordered Pb layer prepared on Si(111), which besides being stable up to 450 K is inert in ultrahigh vacuum (UHV).

The experiments have been carried out in three UHV chambers with base pressures in the low 10^{-10} Torr. The first one contains a variable temperature STM. The second chamber is a high-resolution helium atom scattering (HAS) apparatus with a time-of-flight arm and a fixed angle of 105.4° between incident and outgoing beam,[11] whereas the third chamber is a HAS apparatus which enables determination of absolute diffraction reflectivities.[4,8] In this system, the incident beam's intensity can be measured and used to normalize scattered beam intensities with respect to the incident beam, thereby yielding absolute diffraction intensities.

The three chambers offer the capability to evaporate *in situ*, a rear view low energy electron diffraction (LEED) optics that is also used for auger electron spectroscopy (AES), ion gun and mass spectrometer. We have used as substrates high-quality Si(111) wafers, 0.5 mm thick, that were cleaned by standard methods prior to insertion in the UHV chambers. Inside the vacuum the samples were cleaned by heating to 1400 K while keeping the base pressure in the low 10^{-10} Torr regime, which led to the appearance of excellent He diffraction patterns from the (7×7) surface reconstruction of Si(111). STM examination of the clean surfaces shows atomically resolved terraces larger than 2 microns, confirming the very low misalignment of the wafers. Pb was evaporated from Knudsen cells at slow rates of 0.1–0.7 ML/min, while the samples were either in the microscope or in the He diffractometers at 90–150 K. In the HAS experi-

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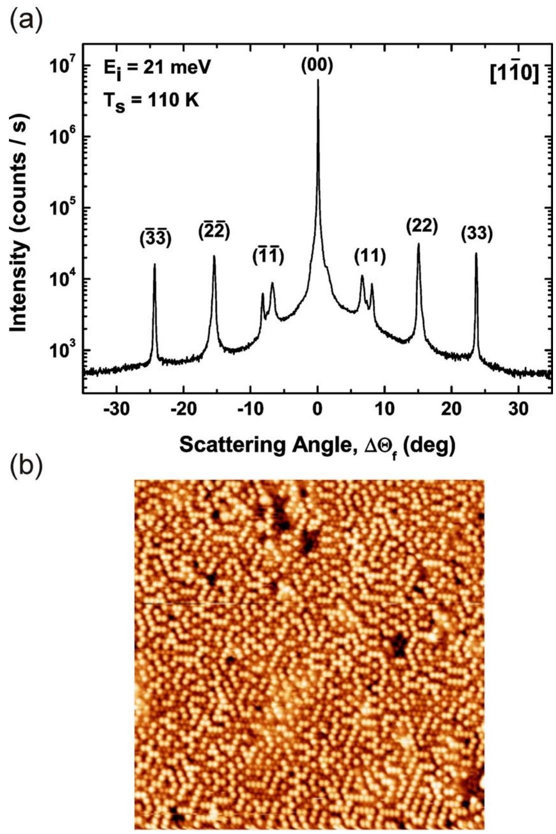

FIG. 1. (Color online) (a) Angular distribution of He scattered from the Pb($\sqrt{3}\times\sqrt{3}$)R30/Si(111) surface along the [1$\bar{1}$0] azimuth. (b) 50 $\times$50 nm^2 STM image of the surface measured with a bias voltage of -1.5 V and a tunneling current of 1.0 nA.

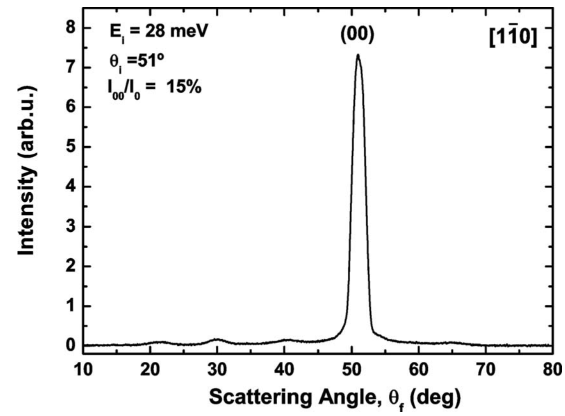

FIG. 2. He-diffraction spectrum from the Pb($\sqrt{3}\times\sqrt{3}$)R30/Si(111) surface along the [1$\bar{1}$0] azimuth measured with the set up used to perform absolute intensity calibration. The surface temperature is 110 K.

ments, Pb coverage calibration was done by monitoring the specular He intensity during Pb deposition at 320 K, which exhibits a well defined maximum at a coverage of 1/3 ML, i.e., when the well-ordered ($\sqrt{3}\times\sqrt{3}$)R30 structure is completed. Because of the low deposition rate used, the error in the coverage determination is ± 0.02 ML. Further details on coverage calibration are given elsewhere.[8] For the experiments reported here, the Pb($\sqrt{3}\times\sqrt{3}$)R30 structure was prepared by depositing 3 MLs of Pb at 100 K and heating to 700 K, which desorbs the excess Pb.

Figure 1 shows the He-diffraction pattern obtained for the Pb($\sqrt{3}\times\sqrt{3}$)R30 structure, measured in the fixed angle set-up along the [1$\bar{1}$0] azimuth. Although specular scattering dominates, diffraction up to third order is clearly resolved in the spectrum. The existence of Pb islands with other periodicities, mainly Pb($\sqrt{7}\times\sqrt{3}$),[12–14] leads to the appearance of the small double peaks present at approximately $\pm 8°$ from the specular peak. It is worth mentioning that the corresponding intensity is almost three orders of magnitude smaller than that of the specular peak, which is the one of interest for Helium Atom Microscopy.

The corresponding STM image measured at negative bias voltage is also shown in Fig. 1. Depending on the annealing temperature the Pb($\sqrt{3}\times\sqrt{3}$)R30 presents a number of defects, most of them are substitutional Si atoms which displays a strong bias voltage dependency.[15]

In order to determine the absolute reflectivity of this sample, the experiment was repeated using the second HAS apparatus. The He-diffraction spectrum, recorded at an incident energy of 28 meV, is shown in Fig. 2. Also here specular diffraction dominates, and small diffraction peaks up to third order are also visible. Under these conditions, 15% of the incoming beam intensity is scattered into the specular peak. This is a factor of $\sim$20 times higher than for Si(111)-H(1$\times$1) passivated surfaces under similar scattering conditions,[16] and comparable with the reflectivity of the QSE-stabilized Pb thin films on Si(111).[8] However, and in contrast to the latter—which are stable only at surface temperatures below 250 K-Pb($\sqrt{3}\times\sqrt{3}$)R30 surfaces are stable up to 450 K, which is a great advantage for application purposes.

These results demonstrate that a Pb($\sqrt{3}\times\sqrt{3}$)R30 structure exhibits a high-reflectivity for He atoms. A He specular reflectivity of 15% can be obtained from these surfaces at 110 K, working at $\theta_i=51°$ and $E_i=28$ meV. In order to use such films as a focusing mirror for a scanning helium atom microscope, it is convenient to analyze under which conditions this value could be further improved.

He atom beams scattered from a solid surface are attenuated according to the so-called Debye–Waller model. In this model, the intensity $I(T)$ of a diffraction peak is related to the intensity I_0 from a lattice at 0 K by as follows:

$$I(T) = I_0 e^{-2W(T)}, \quad (1)$$

where $\exp[-2W(T)]$ is the Debye–Waller factor. When specular reflection is considered, $W(T)$ can be put as a function of the incident beam energy E_i and the angle of incidence θ_i as follows:[4]

$$W(T) = 12m(E_i \cos^2 \theta_i + D)T/Mk_B\Theta_D^2, \quad (2)$$

where M is the mass of a surface atom, m is the mass of the incoming particle, k_B is the Boltzmann constant, Θ_D is the surface Debye temperature, and D is the potential well depth.

Figure 3 shows the dependence of specular intensity for the Pb($\sqrt{3}\times\sqrt{3}$)R30 surface as a function of both surface temperature (a) and incident beam energy (b). For comparison, the dependence obtained for the clean Si(111)-(7$\times$7) structure is also shown in Fig. 3(a). We see that the absolute values obtained for the Pb($\sqrt{3}\times\sqrt{3}$)R30 surface are a factor of $\sim$100 higher than for the clean surface, which means that in this temperature range, the Pb covered surface behaves as a harder material, with a corresponding larger Θ_D. The steep slope exhibited by this surface as a function of surface tem-

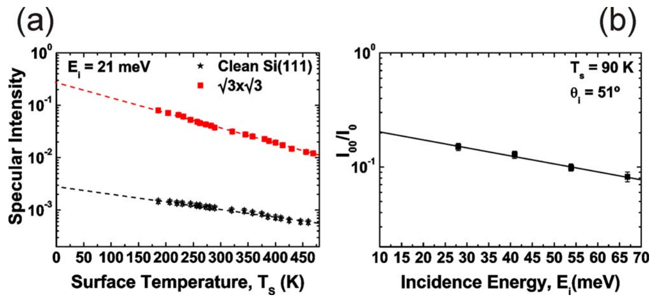

FIG. 3. (Color online) Specular intensity dependence for the Pb($\sqrt{3} \times \sqrt{3}$)R30/Si(111) surface as a function of surface temperature (a) and incident beam energy (b).

perature, combined with the dependence observed with incident beam energy, suggests that the absolute specular reflectivity could be increased to $\sim$40% by combining a large angle of incidence ($\theta_i \sim 70°$) with an incidence energy close to $E_i \sim 10$ meV, while keeping the surface at 50 K. As a consequence, scanning helium atom microscopes designed to use these surfaces as mirrors could get a signal to noise ratio several orders of magnitude larger than current prototypes, which would allow a significant improvement in the lateral resolution achievable.

Finally, we have checked that the reflectivity of Pb thin films remains almost unchanged in a time scale of several weeks. Figure 4(a) shows a He diffraction pattern recorded for a Pb($\sqrt{3} \times \sqrt{3}$)R30 surface, stored four weeks in UHV (blue curve). This spectrum is almost identical to the one shown in Fig. 1 (measured immediately after preparation of the surface), the major change is just a decrease in specular reflectivity of less than 10%. This means that the sticking probability of the residual gases present in UHV (mainly hydrogen and water) on these surfaces is very low. The original spectrum can be recovered by flash annealing the sample to 450 K, as illustrated by the black curve.

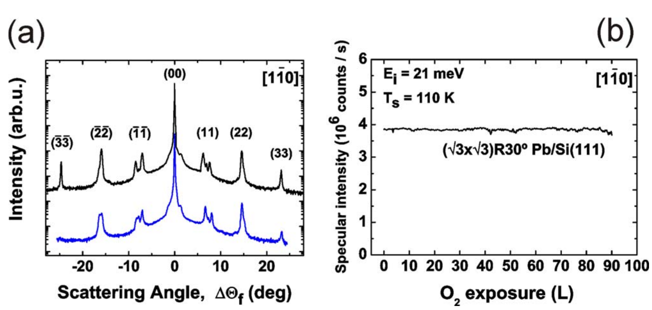

FIG. 4. (Color online) (a) He-diffraction spectra measured for a Pb($\sqrt{3} \times \sqrt{3}$)R30/Si(111) surface after four weeks in UHV (bottom curve) and after applying a flash annealing to 450 K (top curve). The curves have been shifted with respect to each other to allow a better comparison between them. (b) He specular intensity from this surface as a function of oxygen exposure.

We have complemented these results by studying the surface reactivity in more detail. This was done by exposing the Pb($\sqrt{3} \times \sqrt{3}$)R30 surface to molecular oxygen at a partial pressure of 2×10^{-7} mbar, while monitoring the specular peak intensity. The result of this experiment is also shown in Fig. 4(b), and reveals that the specular intensity decreases by ca. 10% after an exposure time of 600 s, which corresponds to a total dose of 90 Langmuir. Although we have observed that the surface is contaminated when exposed to air, these results demonstrate that this surface is very inert while in UHV, making it an ideal candidates to be used as a mirror in scanning helium atom microscopes.

In conclusion, we have shown that a Pb($\sqrt{3} \times \sqrt{3}$)R30/Si(111) surface is an excellent mirror for He atoms, with an absolute specular reflectivity of 15%. Such surfaces are stable up to 450 K, and their reflectivities remain unchanged in UHV in a time scale of several weeks.

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