

A Quantum-Stabilized Mirror for Atoms**

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Helium atom scattering (HAS) is a powerful, well-established technique for investigating the structural and dynamical properties of surfaces.^[1,2] Because of the low energies used (ca. 100 meV), neutral He atoms probe the top-most surface layer of any material in an inert, completely nondestructive manner. This means that a scanning helium atom microscope using a focused beam of He atoms as imaging probe would be a unique tool for reflection or transmission microscopy, with a potential lateral resolution of ca. 50 nm. It could be used to investigate insulating glass surfaces, delicate biological materials, and fragile samples, which are difficult to examine by other methods because of sample charging or electron excitation effects. The practical realization of such a microscope requires the development of a mirror that is able to focus a beam of low-energy He atoms into a small spot on the sample to be examined. Holst and Allison^[3] 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 μm. 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%.

Here we show that quantum size effects (QSEs) can be exploited to produce an ultraperfect, atomically flat film of Pb of “magic” thickness on a highly perfect Si(111) thin wafer,

where magic refers to certain thicknesses that are more energetically favored than others. The metal film reproduces the structural perfection of the substrate, is atomically flat over micrometer-wide areas, and stable up to 250 K. As a consequence, more than 15% of the incoming He atoms are scattered from this quantum-stabilized surface in the specular direction, which allows its use as an ultrasmooth mirror for neutral atoms; a device of interest also for atom optics in order to manipulate matter waves coherently.^[4]

Finding materials suitable for atom lenses or mirrors is a daunting task. Although the combination of microskimmers with Fresnel zone plates was demonstrated to focus a He beam down to 1.5 μm^[5] and even though the first real two-dimensional He-microscopy images with a similar resolution have recently been published,^[6] the very low intensity in the focused specular peak poses a serious limitation to developing an efficient atom microscope. Mirror focusing has significant advantages compared to zone plates: numerical apertures can be much larger, and it offers true *white light* focusing, with no chromatic aberrations.

The problems of using a surface for atom optics must be considered at both the macroscopic level, where classical mechanics is applicable, and the microscopic level, where quantum effects dominate. At the macroscopic scale, the mirror must be bent to a Cartesian reflector surface to avoid aberrations, which limits the maximum surface deviation resulting from bow or thickness variations to ±0.5 μm.^[7] The bending can be achieved electrostatically, which requires the mirror to be ultrathin. Recent results have shown that 50 μm thick Si(111) wafers with such properties can be produced by improving current crystal-cut and -polishing technologies.^[8]

The most serious problem arises at the microscopic scale, owing to the high sensitivity to surface defects characteristic of He atom scattering.^[9] This requires surfaces of outstanding crystalline perfection, homogeneous over lateral distances larger than the coherence length (ca. 250 Å) and chemically inert, because the mirror surface must be held atomically clean for long periods.

The surfaces of ultrathin semiconductor crystals can be produced with smaller density of steps and point defects than metal surfaces, but the larger charge corrugation at semiconductor surfaces results in a severe loss of intensity from the specular beam into diffracted beams, resulting in specular intensities of the order of less than ca. 1% of the incident beam. Metals, the compact surfaces of which reflect He atoms mostly into the specular beam^[2] with high reflectivities (ca. 25%), can not be bent reproducibly. Thus, the optimum reflecting surface

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for a He atom mirror could consist of a 50 μm thick, ultraperfect semiconductor wafer covered by a metal film of nanometer thickness, provided that the latter will uniformly cover the semiconductor surface at the atomic scale.

Unfortunately, many metals show a persistent tendency to grow on most semiconducting surfaces forming 3D islands with a broad distribution of heights. Recently, however, it has been found that unusually strong quantum confinement of the electrons in some ultrathin metal films deposited on substrates with an absolute (or symmetry) gap around the Fermi energy, leads to an electronic mode of growth,^[10] which makes certain island heights (“magic”) much more energetically favored than others.^[11–13]

We describe here the exploitation of this QSE^[14,15] to produce a thin film of Pb of magic height on a particularly perfect Si(111) thin wafer. The film is atomically smooth over micrometer-wide areas. We report on He scattering from this quantum-stabilized surface and demonstrate that the incident beam is scattered only into the specular beam with a reflectivity comparable with the best metallic crystals, allowing its use as a quantum-stabilized mirror for atoms.

The experiments have been carried out in three different ultrahigh-vacuum (UHV) chambers with base pressures in the low 10^{-10} Torr range ($1 \text{ Torr} = 1.333 \times 10^2 \text{ Pa}$). The first one contained a variable temperature scanning tunneling microscopy (STM) instrument. The second chamber was a high-resolution He scattering apparatus with a time-of-flight arm and a fixed angle of 105.4° between the incident and outgoing beam,^[16] whereas the third chamber was a He scattering apparatus that enabled determination of absolute diffraction reflectivities (see Experimental). 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 high-quality Si(111) wafers as substrates, 0.5 mm thick, which 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), as shown in the top panel of Figure 1. STM examination of the clean surfaces shows atomically resolved terraces larger than $2 \mu\text{m}$, confirming the very low misalignment of the wafers. Pb was evaporated from Knudsen cells at slow rates of $0.1\text{--}0.7 \text{ monolayer (ML) min}^{-1}$, while the samples were either in the microscope or in the He diffractometers at $90\text{--}150 \text{ K}$. Details on coverage calibration are given elsewhere (see Experimental section).

Figure 1 (bottom) shows the specular intensity of He measured during the deposition of Pb on Si(111)- (7×7) at 140 K. The initial specular intensity from Si(111)- (7×7) is fairly small ($7.5 \times 10^4 \text{ counts s}^{-1}$), as most of the intensity goes into the numerous diffracted beams (see Fig. 1, top). Apart from a first maximum upon completion of the wetting layer, the specularly reflected intensity is negligible for the first 4 MLs of Pb. It reaches a maximum 25 times more intense than on the

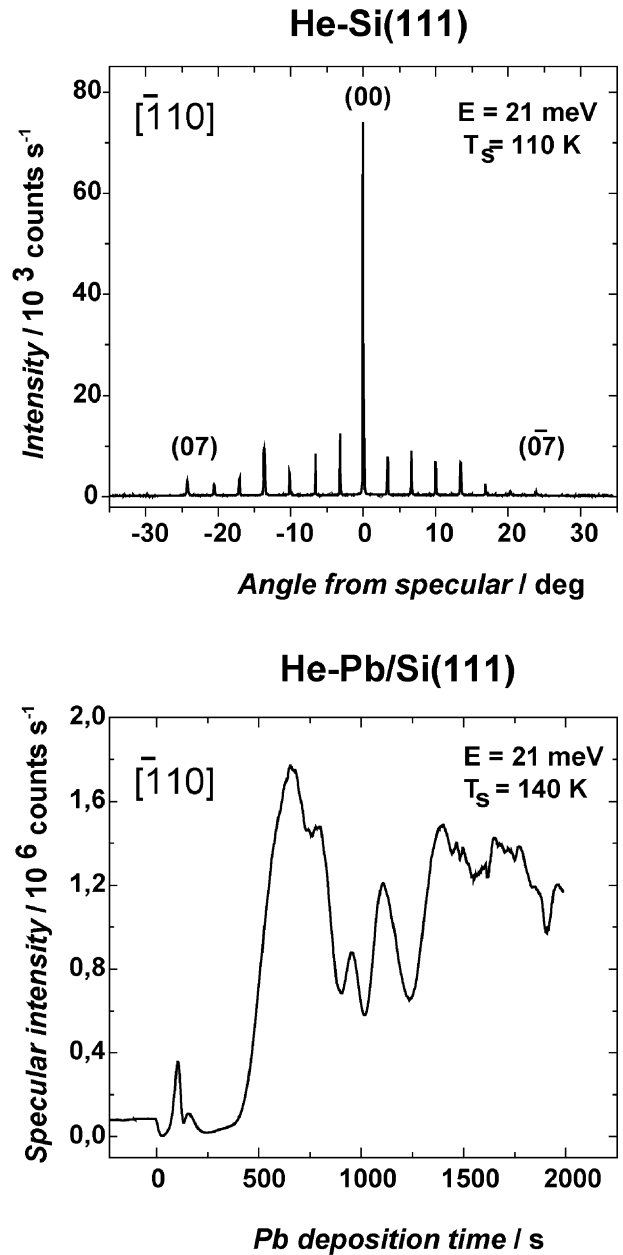


Figure 1. Top: Angular scans of He scattered from the clean Si(111)- (7×7) surface along the $[1\text{--}10]$ azimuth. Bottom: Evolution of the He specular intensity during the deposition of Pb on top of this surface at 140 K.

starting surface for a 4 ML thick Pb film, which corresponds to the completion of the flat-top, magic-height islands that are the first stage of the growth.^[17] From that moment on, the intensity oscillates as new Pb layers are added, initially in a layer-by-layer fashion, followed by bilayer growth.^[18,19]

STM imaging shows that growing directly on the Si(111)- (7×7) substrate does not result in Pb films of enough lateral perfection or thermal stability.^[20] If the starting surface is prepared, however, to present a $(\sqrt{3} \times \sqrt{3})R30$ Pb-induced

surface reconstruction,^[21] subsequent deposition of Pb leads to films of much higher perfection. Figure 2 shows a series of STM images recorded for increasing Pb coverages during deposition on the $(\sqrt{3} \times \sqrt{3})R30$ Pb-induced reconstruction surface at 160 K, which are characteristic of the surface morphology in the 100–160 K range. Figure 2a corresponds to a deposition of 0.7 ML of Pb and shows the presence of a dense array of small features 2 Å high that uniformly cover the substrate. This is the dense $(\sqrt{3} \times \sqrt{3})$ phase that acts as a wetting layer for further growth. It corresponds to 2/3 of a ML of Pb. After deposition of 1.3 MLs of Pb (Fig. 2b), the surface consists of small islands with a size that has increased with the coverage, but still remains smaller than the coherence length of the incoming He beam. The large density of island boundaries and defects is consistent with the reduced reflected intensity of He before the first maximum at 2 MLs.

Figure 2c corresponds to a deposited thickness of 6.6 MLs and shows that, by the time the specular intensity displays clear oscillations, the surface contains islands, typically 250 Å wide. The Pb islands show a (111)-oriented atomically flat top, as demonstrated by atomic resolution images (not shown). Most

of the islands have the same height and the others are 1 ML higher.

Figure 2d shows the specular intensity recorded during deposition of Pb on $(\sqrt{3} \times \sqrt{3})R30$ Pb-induced reconstruction surface at 140 K. Well-defined oscillations were detected in a wide temperature range (100–160 K). There is a hint of a first maximum after completion of the wetting layer, followed by clear maxima that are separated by the time needed to deposit a single monolayer from 2 to 7 MLs. From 10 MLs on, maxima in the specular intensity are observed every 2 MLs. The overall behavior of the reflected He intensity is similar to the one reported for the deposition of Pb on Cu(111) at 140 K.^[15]

The maxima in the reflected He specular intensity arise from the fact that during the growth, Pb films of certain atomic heights are energetically much more stable than others, leading to islands of specific, magic heights.^[11–13] Each island is a (111)-oriented Pb nanocrystallite, where electrons from the sp band of Pb are efficiently confined between the vacuum barrier and the Si band gap around the Fermi energy. The confinement discretizes the s-p band of Pb and the corresponding quantum well states (QWSs) can be detected by local tunneling spectroscopy^[13] or angular-resolved photoelectron spectroscopy.^[12] The sequential population of the QWSs leads to transport,^[22] superconducting,^[23,24] thermodynamic stability,^[25,26] or reactive^[27] properties that oscillate in magnitude with the thickness of the film.

Films deposited at low temperature are continuous, but atomically rough, as they usually contain significant fractions of (at least) three adjacent atomic thicknesses, that is, an 8 ML thick film deposited at 136 K (not shown) consists of 25.4% of the area with 9 MLs, 39% of 8 MLs, and 36.5% of 7 MLs (always above the wetting layer). STM at variable temperature is used to follow the evolution of the morphology of the films during quasi-static annealing with temperature ramps of 1 K min^{-1} . The thermal stability and, thus, the suitability to act as an atom mirror depends on whether the deposited Pb film has a magic or nonmagic thickness. If the film has a noninteger coverage in monolayers, voids and pits also appear upon annealing. Figure 3 shows selected, $0.5 \mu\text{m}$ wide snapshots from an STM movie recorded during the heating of the 6.6 ML Pb film shown in Figure 2. The film has a flat, granular structure with no change up to 180 K. Above 200 K, the dominant height (6 MLs) start to decompose and pits that reach down to the wetting layer appear. The image at 268 K illustrates the decomposition of the film

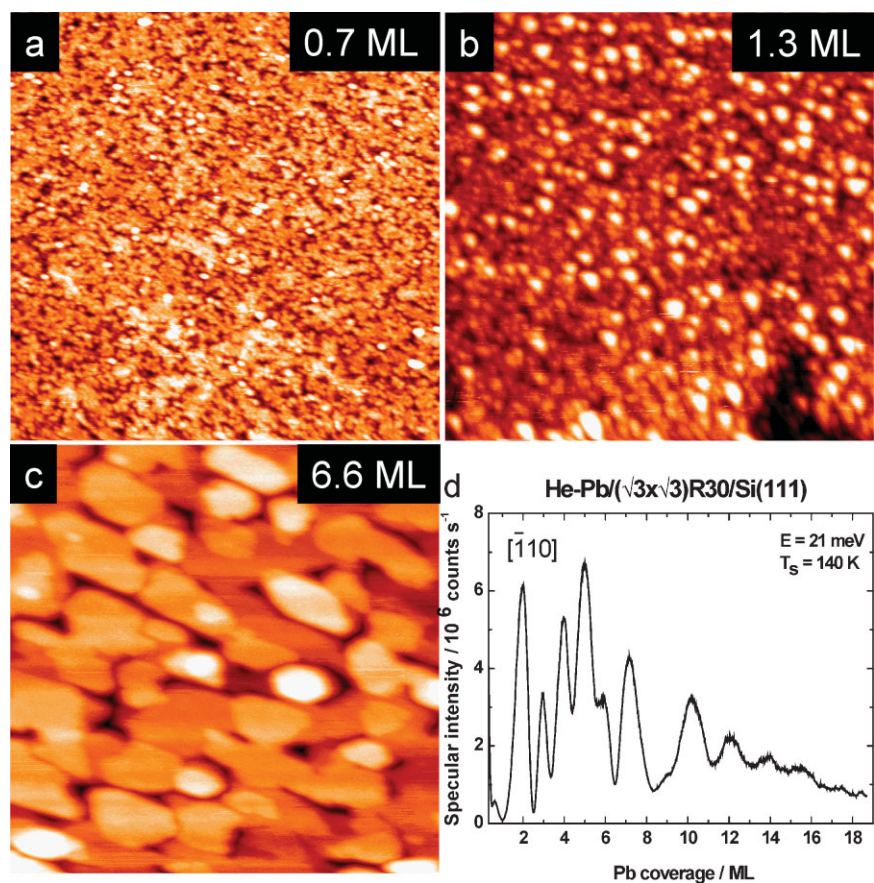


Figure 2. a–c) $100 \times 100 \text{ nm}^2$ STM images recorded during the deposition of Pb on the $(\sqrt{3} \times \sqrt{3})R30/Si(111)$ surface at 160 K. The images have been taken with a sample bias of 3 V and correspond to 0.7 (a), 1.3 (b), and 6.6 MLs (c) of additional Pb. d) Evolution of the specularly reflected He beam intensity during the deposition of Pb on top of a $(\sqrt{3} \times \sqrt{3})R30/Si(111)$ surface at 140 K.

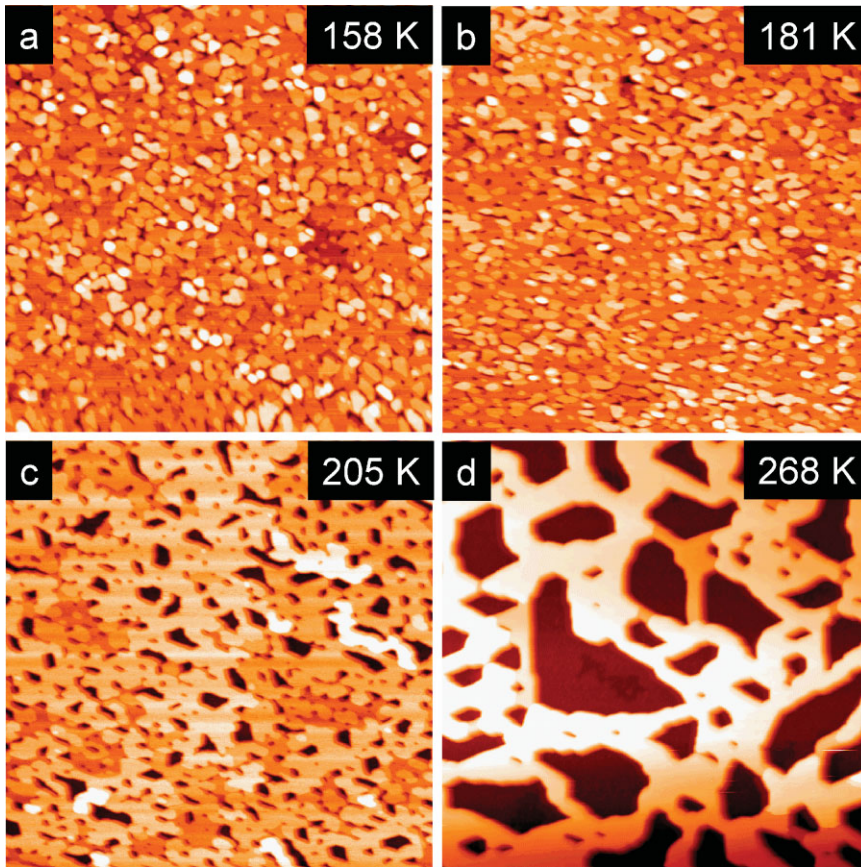


Figure 3. Series of $500 \times 500 \text{ nm}^2$ STM images of a 6.6 ML thick Pb film deposited at 160 K and heated to different temperatures. Not a single substrate step is visible in the image.

in regions 9, 11, and 13 ML high, that is, some of the magic thicknesses. The dark areas correspond to the wetting layer.

Heating Pb films with a coverage close to completion of one of the magic thicknesses, on the contrary, increases their structural perfection and results in a further increase in the He intensity specularly reflected, as larger areas of the films become atomically flat. Figure 4 shows the surface morphology of a 7.1 ML thick film of Pb deposited at 114 K and annealed to 260 K. The film is atomically flat and most of the film (94% of the surface) is 7 MLs thick. Only 5% of the surface is occupied by 9 ML thick regions (bright areas) and ca. 1% by 5 ML thick regions, imaged as dark small islands. Notice that not a single substrate step is visible in the image. Very-large-scale STM images indicate that the film at 260 K is atomically flat over lateral scales larger than $10 \mu\text{m}$.

For these films, the specular reflectivity is 15% of the incident He beam, that is, 15–20 times higher than for Si(111)-H(1×1) passivated surfaces under similar scattering conditions^[28] and comparable with the reflectivity of the best metallic single crystals.^[2] The He diffraction spectrum recorded along the $[-110]$ azimuth is shown in Figure 4 (bottom). Note that only specular diffraction is observed in the angular distribution, as expected from a close-packed metal surface. Because all magic thicknesses achieve similar

structural perfection upon annealing, the maximum reflected intensity is approximately the same upon heating to the temperature range in which each of them are stable.^[18,20,25]

A practical requirement for using these surfaces as a focusing mirror for microscopy is long-term stability in UHV. Pb thin films of magic height are stable in UHV during weeks if kept below 200 K, as well as inert to oxygen exposure up to pressures of the order of 10^{-7} Torr, even at a surface temperature of 90 K (see Experimental section). Films of nonmagic thickness, on the contrary, are reactive and unstable. In fact, increased stability^[25,26] and reduced chemical reactivity^[27] are both related to the lack of QWSs close to the Fermi level and the corresponding decreased density of states for magic thicknesses.

Our results show that a quantum-controlled mode of growth driven by electron confinement can be used to stabilize atomically flat, ultrathin films of Pb on Si(111) close to room temperature, with the same degree of surface structural perfection as the substrate. A He specular reflectivity of 15% can be obtained from these surfaces at 110 K, working at $\theta_i = 51^\circ$ and $E_i = 28 \text{ meV}$. 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 $I_k(T)$ of a diffraction peak with momentum transfer $\hbar k = \hbar(k_i - k_f)$ at a crystal temperature T is given by

$$I_k(T) = I_k(0)e^{-2W(T)} \quad (1)$$

where $I_k(0)$ is the diffracted intensity at 0 K surface temperature and $W(T)$ is the Debye–Waller exponent. For the specular peak, it can be expressed as a function of the incident beam energy E_i and the angle of incidence θ_i ^[2]

$$W(T) = 12m(E_i \cos^2 \theta_i + D)T / M k_B \theta_D^2 \quad (2)$$

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

Equation 2 provides a hint of why, at a given surface temperature, a much larger He reflectivity is expected from surfaces containing heavier atoms (such as Pb) as compared to

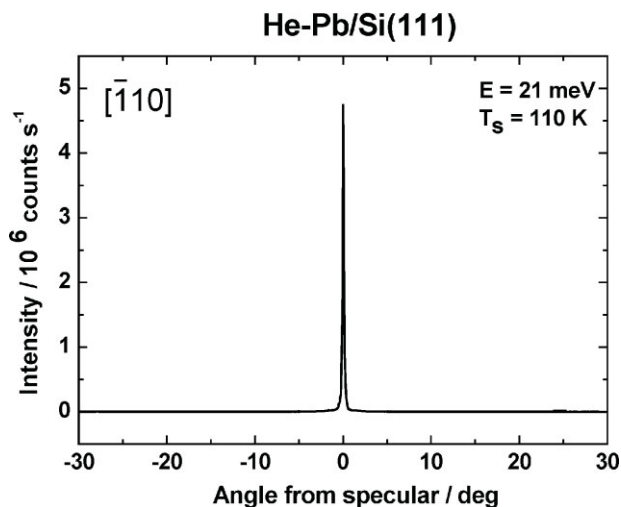
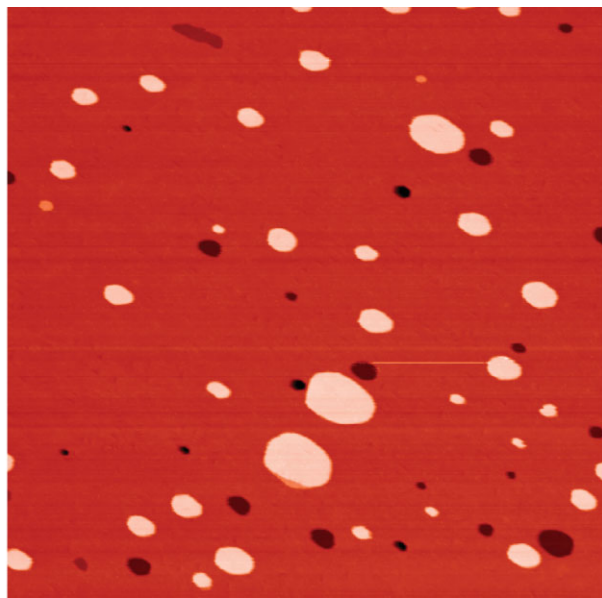


Figure 4. Top: $500 \times 500 \text{ nm}^2$ STM image of 7 ML thick Pb film deposited at 98 K and heated to 260 K. Most (94%) of the surface is covered with 7 MLs. Bottom: He diffraction spectrum corresponding to a surface covered with 4 MLs of Pb at 110 K.

light ones [such as a H-passivated Si(111) surface]. It also indicates that the intensity of a specularly reflected beam should be highest for large angles of incidence and small incidence energy. Therefore, specular reflectivity values as large as ca. 40% could be obtained by combining a large angle of incidence ($\theta_i \sim 70^\circ$) with an incidence energy close to $E_i \sim 10 \text{ meV}$, while keeping the surface at 50 K. This means that a scanning helium atom microscope designed to work under these conditions and using a quantum-stabilized focusing mirror might have a signal several orders of magnitude larger than current prototypes, which would allow to enhance accordingly the lateral resolution achievable.

In summary, we have shown that QSEs can be used to stabilize atomically flat Pb films close to room temperature

with the same degree of perfection as the Si(111) substrate. The very high He atom reflectivity observed suits them to act as quantum-stabilized, ultrasmooth, focusing mirrors for neutral atoms.

Experimental

Determination of Absolute He Diffraction Reflectivities: With the molecular beam apparatus used in our experiments, the intensity of the incident beam could also be measured and used to normalize scattered beam intensities with respect to the incident beam, thereby yielding absolute diffraction intensities. Because the incident and specular beams had similar widths, this normalization could be made just by comparing the areas of the incident and specular peaks for a given incidence condition.

Sample Stability: We have checked that the reflectivity of Pb thin film remains almost unchanged on a time scale of several weeks. In our experiments, we observed a decrease in specular reflectivity of less than 10% after four weeks storage in UHV. This means that the sticking probability of the molecules present in UHV (mainly hydrogen and water) on these surfaces was very low, which we also studied by performing oxygen adsorption experiments in UHV. In these experiments, Pb thin films of different heights, kept at 90 K, were exposed to molecular oxygen at a pressure $p = 2 \times 10^{-7}$ Torr while monitoring the specular peak. We observed that the specular intensity decreased by ca. 10% after an exposure time of 600 s, which corresponds to a total dose of 120 Langmuir.

Pb Coverage Calibration: The experiments were carried out in three different ultrahigh-vacuum (UHV) chambers. Two of them were HAS machines, and the third one contained a variable-temperature STM. All three chambers had the capability to evaporate in situ Pb from a Knudsen cell on the Si(111) substrate. The calibration of the Knudsen cell was crucial for these experiments, because the amount of Pb deposited on the surface dictates its properties. In the HAS experiments, this was done by monitoring the specular He intensity during Pb deposition at 320 K, which exhibited a well-defined maximum at a coverage of 1/3 ML, that is, when the well-ordered $(\sqrt{3} \times \sqrt{3})R30$ structure was completed. Because of the low deposition rate used, the error in the coverage determination was ± 0.02 ML.

In the STM measurements, we used two different methods to calibrate and cross-check the amount of Pb deposited on the Si(111) surface. In the first method, Pb was deposited at room temperature on the Si(111) surface. It is well-known that under these conditions Pb grows in the Stranski–Krastanov mode. To determine the amount of Pb deposited, we measured the height and lateral size of the Pb islands present on the surface. This method had the disadvantage that large scan areas are needed to make sure that the studied area is representative of the surface, because the 3D islands are nucleated every $0.5 \mu\text{m}$ on average.

The second method consisted in evaporating Pb with the sample held at 160 K. Under these conditions the growth mode changed to layer-by-layer, and the surface was completely covered by Pb. Once the evaporation was finished, we slowly changed the temperature of the STM and measured STM images every 5 K. At a certain temperature, the Pb films broke apart and formed 3D islands, leaving the so-called wetting layer between the islands. This layer consisted of 1/3 of a Pb monolayer alloyed with Si and formed a $(\sqrt{3} \times \sqrt{3})R30$ structure that was very easy to identify with atomic resolved images. Once the film broke apart, we could measure the height and size of the islands and calculated the amount of Pb originally deposited onto the surface.

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