Flexible thin metal crystals as focusing mirrors for neutral atomic beams

Gloria Anemone,¹ Amjad Al Taleb,¹ Sabrina D. Eder,² Bodil Holst,² and Daniel Farías^{1,3,4}

¹Departamento de Física de la Materia Condensada, Universidad Autónoma de Madrid, 28049 Madrid, Spain

²Department of Physics and Technology, University of Bergen, Allègaten 55, 5007 Bergen, Norway

³Instituto "Nicolás Cabrera," Universidad Autónoma de Madrid, 28049 Madrid, Spain

⁴Condensed Matter Physics Center (IFIMAC), Universidad Autónoma de Madrid, 28049 Madrid, Spain

(Received 9 March 2017; published 22 May 2017)

The development of novel reflective optical elements is essential to improve the focusing of neutral atomic beams. The recent availability of commercial thin crystals led to a renewed interest in curved mirrors as reflective elements for He microscopy. We have investigated the reflectivity to incoming He atoms of Cu(111), Ni(111), and Ru(0001) crystals of thickness between 50 and 150 μ m. The results have been compared with the ones obtained from bulk crystal surfaces. Our study reveals that a 100 μ m thick Cu(111) crystal is the best candidate to be employed as a curved mirror, with an absolute reflectivity of 20% and a long-range crystalline order larger than 200 nm. In contrast, much lower reflectivities (3%–14%) have been measured for thin Ni(111) and Ru(0001) crystals. Finally, we show that a thin (100 μ m) Cu(111) crystal can be bent by an electrostatic field to focus an incoming He beam to a spot of 350 μ m. Due to the focusing properties of the mirror, a direct beam with less collimation can be used, leading to a larger reflected intensity. The absolute focused intensity is two orders of magnitude larger than previously reported. This represents a big step forward towards achieving the goal of building a high-resolution scanning helium atom microscope.

DOI: 10.1103/PhysRevB.95.205428

I. INTRODUCTION

Thermal energy neutral helium atoms probe the outermost surface layer of any material in an inert, completely nondestructive manner [1]. Due to the low energies used (usually less than 100 meV), the interaction occurs through the electron density distribution of the topmost surface atoms. Thus, He atoms are ideally suited to investigate all kinds of materials, including insulating and/or fragile surfaces, biological samples, or polymeric nanostructures, without damaging them. One of the crucial challenges for the construction of high-resolution He microscopes is the development of optical elements that can handle the focusing of neutral He atoms.

In order to solve this problem, two main paths have been pursued in the last decade: diffractive optical elements using Fresnel zone plates [2–4], and reflective elements using curved mirrors [5,6]. By using Fresnel zone plates, the helium beam has been focused below 1 μ m [3]. However, a major problem with Fresnel zone plates is that they suffer from chromatic aberrations, in addition to the low intensity of the focused beam (approximately 5%–10% of the incident beam). Chromatic aberration is an issue for He microscopy, because the He beam, generated by a supersonic expansion, will always have a given velocity distribution [7,8]. The use of curved mirror surfaces could solve these limitations. Besides being inherently achromatic, mirrors can focus an incident beam with a diameter of several millimeters, leading to higher focused beam intensities.

Creating surfaces that are both curved and smooth at the atomic scale is a major challenge. Due to the high sensitivity of He atoms to surface defects, their density must be very low [9]. A good candidate to be used as a mirror is a thin semiconductor crystal, like Si(111), since their surfaces can be manufactured with larger terraces and smaller density of defects than metal surfaces. This approach has been followed in the past, using

electrostatically bent thin Si(111) crystals as mirrors. However, two main problems persist when semiconductor crystals are used: the poor flexibility and the large surface corrugation compared to metal crystals. The former is essential to bend the crystal electrostatically, whereas the latter imposes a limitation to the intensity of the specular peak. In effect, highly corrugated surfaces lead to the appearance of many diffraction channels, causing a loss in intensity of the specular peak [1]. As a consequence, the absolute specular reflectivity for Si(111) is with approximately 1% considerably smaller than the high reflectivity measured for metal surfaces (from 15% to 40%) [10,11].

Until now, technical restrictions limited the thickness of the metal crystals used. However, nowadays commercial crystals of about 50 μ m are available for many metals. This characteristic allows the manipulation of metallic surfaces and the possibility of a controlled bending of the metal itself. But so far no investigations have been reported on the surface quality and the long-range order of these thin crystals, which are determinant factors of the quality of the reflected He beam.

We have studied four different thin crystal surfaces: Cu(111) of 100 μ m, Ni(111) of 100 and 50 μ m, and Ru(0001) of 150 μ m. In order to asses the quality of these surfaces, these results have been compared with data measured from bulk crystals of the same metals. These metals are especially interesting from the point of view of their use as mirrors for He atoms, since a graphene layer can be grown on Ru(0001), Ni(111), and Cu(111). This keeps the surface clean and stable even after exposure to ambient conditions, while the absolute reflectivity remains high: more than 20% for graphene/Ru(0001) [12,13] and graphene/Ni(111) [11,14], and 5% for graphene/Cu(111) [15,16]; in addition, the surfaces maintain their reflectivity in vacuum for periods of weeks or month, a huge advantage for microscopy applications. In our current study, we demonstrate that a thin (100 μ m) Cu(111) crystal exhibits the largest ordered domains, and that its flexibility allows focusing He atoms with a very high reflectivity.

II. EXPERIMENTAL DETAILS

Three different thin metal crystals from MaTecK have been used in this study: Cu(111), Ni(111), and Ru(0001). The crystals are disks with a diameter of 10 mm and with different thicknesses: 100 μ m for Cu(111), 100 and 50 μ m for Ni(111), and 150 μ m for Ru(0001). A comparison has been made with results obtained with Cu(111), Ni(111), and Ru(0001) bulk crystals. These samples are 2 mm thick, and have disk shapes, with a diameter of 8 mm.

The crystals were mounted on a sample holder which can be heated by electronic bombardment and cooled down to 100 K using liquid nitrogen. The surface temperature was measured with a C type thermocouple spot welded to the sample edge. For the thin crystals, the mounting has been modified to prevent a deformation of the thin crystals themselves. For this purpose, the sample has been placed between a molybdenum disk at the bottom and a molybdenum ring with an aperture of 5 mm on top, and tightened so that the sample remains as flat as possible during the preparation procedure. The electrostatic bending of the sample was achieved by removing the molybdenum disk, allowing the free movement of the thin crystals. The sample, electrically grounded with the upper electrode (the molybdenum ring), remained insulated from the lower electrode (the filament) by a sapphire ring, allowing high voltage application to the electrode structure.

Clean metal surfaces were prepared *in situ* in UHV by 15 min cycles of ion sputtering [0.6 keV for Cu(111), 1 keV for Ni(111) and Ru(0001) with $P_{\rm Ar} \simeq 2 \times 10^{-5}$ mbar] followed by flash annealing at approximately 850 K for the Cu(111) and 1170 K for Ru(0001) and Ni(111). Few cycles (2–3) were found to be enough to prepare the thin crystal surfaces, in comparison to more than ten cycles needed for the bulk crystals. Surface cleanliness and order were checked by looking at the angular distribution of the specularly reflected He beam.

Characterization by helium atom scattering (HAS) was carried out using the HAS apparatus located at LASUAM (Laboratorio de Superficies de la Universidad Autónoma de Madrid). The scattered He atoms are recorded by a quadrupole mass spectrometer mounted on a two-axis goniometer. This configuration allows the determination of the absolute diffraction reflectivities by recording directly the incident beam intensity [17]. The incident beam energy can be changed by cooling or heating of the platinum nozzle. The collimation of the incident beam can be varied by a diaphragm whose diameter can be changed through a rotary disk with apertures of different diameters (400–4000 μ m), leading to beam sizes of 1.2 until 12.2 mm, respectively, at the sample position.

III. RESULTS AND DISCUSSION

Figure 1(a) shows a comparison of angular distributions of He atoms scattered from the four thin different metal surfaces: 100 μ m Cu(111), 50 and 100 μ m Ni(111), and 150 μ m Ru(0001). The intensity (*I*) of the specular peaks has been



FIG. 1. (a) Angular distributions of He atoms scattered from different thin crystals: 100 μ m Cu(111) (blue), with a beam energy $E_i = 64$ meV; 50 μ m Ni(111) (green) at $E_i = 64$ meV and 100 μ m Ni(111) (black) at $E_i = 28$ meV, and 150 μ m Ru(0001) (red) at $E_i = 28$ meV. (b) Comparison of He reflection from Cu(111) with two different thicknesses at the same experimental conditions ($E_i = 64$ meV and $T_S = 90$ K): 2 mm crystal (black) and 100 μ m thin crystal (red).

normalized with respect to the intensity of the incident beam (I_0) . Since the intensity and width of the specular peaks depend on the quality of the surface, it is clear that the best result is obtained for the Cu(111) thin crystal (blue). The specular peak of the Cu(111) surface has a FWHM of 1°, which corresponds roughly to the angular resolution of the HAS machine. An estimation of the domain size w is given by the relationship FWHM $\sim 1/w$, where w is known as transfer width that corresponds to the largest period of the grating which can be straightforwardly resolved with a given instrument [7]. Thus, the average terrace size of Cu(111) surface is at least 200 nm, i.e., one order of magnitude larger than the transfer width of our system. The specular absolute reflectivity is 20% for an incident energy $E_i = 64$ meV.

TABLE I. Summary of results obtained for thin and bulk crystals.

Crystal	Thickness	E_i (meV)	Reflectivity	FWHM	Terrace size
Cu(111)	100 µm	64	20%	1°	>200 nm
Ni(111)	$100 \ \mu m$	28	14%	1.7°	49 Å
Ni(111)	$50 \ \mu m$	64	3%	2.5°	20 Å
Ru(0001)	$150 \mu m$	28	5%	1.4°	89 Å
Cu(111)	2 mm	64	11%	1.5°	52 Å
Ni(111)	2 mm	64	43%	1°	>200 nm
Ru(0001)	2 mm	28	53%	1.1°	>200 nm

For the other thin crystals investigated the measured FWHM for the specular peak is larger, and an estimation of the corresponding domain size can be obtained from the surface coherence length [18,19]. The specular peak of the 100 μ m Ni(111) sample (black) has a FWHM of 1.7°, which gives an average terrace size of 5 nm, smaller than the one measured for bulk Ni(111) crystal (>200 nm). Likewise, the absolute specular intensity is 14% at $E_i = 28$ meV, whereas the reflectivity measured for the Ni(111) bulk surface is 43% at the same incident energy. If the thickness of the Ni(111)crystal is reduced, the reflectivity gets worse. A FWHM of 2.5° is measured for the thin Ni(111) crystal of 50 μ m, which yields an average terrace size of 2 nm. Similarly, the reflected specular intensity is reduced to 3% of the incident beam. This effect can be due to two main reasons: the fact that thin metal crystals tend to wrinkle after heating, and the lower quality of thin crystals. The first hypothesis can be excluded since the mounting of the sample forced the crystal to be flat (see Sec. II). Therefore, we believe the second reason is more likely, meaning that a worse surface quality is obtained below a given crystal thickness. Finally, the 150 thin μ m Ru(0001) crystal has been measured (red curve). Also in this case the quality of the surface is not comparable to the bulk one. The FWHM of this thin crystal is 1.4° and the average terrace size is 9 nm. Its specular absolute reflectivity at $E_i = 28$ meV is 5%, which is a factor of 10 smaller than for the Ru(0001) bulk surface under the same incident conditions. A summary of all the measured surfaces is presented in Table I.

The comparison of the spectra in Fig. 1(b) between the thin Cu(111) crystal and the Cu(111) bulk presents a quite remarkable result: the thin Cu(111) crystal shows a specular peak even sharper than the one from the Cu(111) bulk surface (FWHM = 1.5°), measured under similar scattering conditions ($T_S = 90$ K, $E_i = 64$ meV). Furthermore, the absolute specular intensity of the thin crystal is more than twice that of the bulk. These observations demonstrate the high quality of the thin Cu(111) crystal, in particular its long range crystalline order. The high specular reflectivity and large crystalline domains, in addition to its flexibility, makes the 100 μ m Cu(111) crystal surface a good candidate to build a mirror for neutral atomic beams. For this reason, we have selected this surface to investigate its focusing properties.

The Cu(111) surface was mounted using the modified sample holder described in Sec. II. The upper electrode had an aperture of 5 mm to allow the He beam in, and it was electrically grounded. The lower electrode was insulated from



FIG. 2. (a) He diffraction spectrum from Cu(111) thin crystal (black spectrum) and incident beam spectrum (red spectrum), both measured at an incident beam energy $E_i = 64$ meV. Differences in the shape are evident: Whereas the incident beam is Gaussian, the diffracted beam is not. (b) Three Gaussian error functions curve are plotted together with the measured beam profile, each one with a different standard deviation (σ). The error function that best fits the specular profile is the corresponding to $\sigma = 0.3$. This gives an estimated specular size of 350 μ m.

the sample by a sapphire ring, in order to allow applying a high voltage to the electrode structure.

Figure 2(a) shows the best beam focus achieved by reflection obtained with the thin Cu(111) crystal measured at $\theta_i = 60^\circ$ and $E_i = 64$ meV (black), compared to the incident beam (red). The shape of the beam changes when a high voltage is applied to the electrodes: the direct beam is approximately Gaussian, whereas the profile of the reflected beam follows a different distribution. Since the width of the focused beam is smaller than the width of the detector aperture, the measured angular distribution comes out convoluted with the aperture.

In order to estimate the size of the focused beam, we have to take into account how the final distribution is generated under the current experimental conditions. A convolution between a rectangular function (to model the finite aperture of the detector) and a Gaussian (to model the He beam) is used to model the observed angular profile.

For the symmetry of the rectangular function, one may also want to take just a Heaviside function into account. The Heaviside step function

$$f(x) = \begin{cases} 1, & x \ge 0, \\ 0, & x < 0 \end{cases}$$

is the mathematical description for an ideal edge. The Gaussian distribution in one dimension has the form

$$h(x) = \frac{1}{\sigma\sqrt{2\pi}} e^{-\frac{1}{2}(\frac{x-\mu}{\sigma})^2},$$
 (1)

where σ is the standard deviation of the distribution and μ is the mean of the distribution. The best fit for the slope of the observed data is provided by the convolution of a Gaussian with a Heaviside function. It is defined as

$$(f * g) = 1 + \operatorname{erf}\left(\frac{x}{\sqrt{2}\sigma}\right),$$
 (2)

where

$$\operatorname{erf}(x) = \frac{2}{\sqrt{\pi}} \int_0^{+\infty} e^{-\frac{1}{2}(\frac{x-\mu}{\sigma})^2}$$
 (3)

is the error function.

From the best-fit curve shown in Fig. 2(b), we estimate the size of the specular spot to be $350 \ \mu$ m. This is at least a factor 5 better than the values obtained for high-quality flat samples. Unfortunately, due to technical problems with the sample holder, we could not further optimize the focusing conditions, so it is not unlikely that the beam has been focused at a point slightly away from the detector's position. To make sure that the beam has also been focused in the direction perpendicular to the polar scan direction, out-of-plane measurements have been performed (not shown). Also along this direction the width of the specular peak reaches the value of the angular resolution of our equipment it not sufficient to perform an accurate fit along the polar scan direction.

Since we could not change the sample-detector distance, the behavior of the beam at the detector position has been studied by changing the collimation of the incident beam. Five apertures of different diameters (400, 750, 1200, 2000, and 3000 μ m) have been employed to obtain different widths for the direct beam.

Figure 3 shows the FWHM and specular intensity plotted as a function of the beam's size at the diaphragm's position. As expected, the intensity of the incident beam increases with increasing illuminated surface area and the same happens with the specular intensity (red line). The FWHM of the specular peak (blue continuous line), in contrast, remains unchanged even after increasing the illuminated area by a factor of approximately 40, whereas the FWHM of the direct beam increases; the same happens for a flat crystal surface (not shown). In fact, due to the finite aperture dimensions of both



FIG. 3. Effect of beam collimation on the width and the intensity of reflected beam from curved thin 100 μ m Cu(111). Blue: FWHM of reflected and direct beam shown on the left axis. Red: Total intensity of the reflected specular peak shown on the right axis.

detector and source, the angular distributions suffer a certain angular broadening, intrinsic to the instrument [7]. This leads to an increase of the width of the specular peak when the size of the diaphragm aperture increases. For the HAS machine used in the current work, the angular broadening expected for an aperture of 400 μ m is 1.6° (and the one for an aperture of 3000 μ m is 9.6°).

In contrast, for a curved surface focusing the reflected He beam at the detectors position, the angular broadening is expected to be drastically reduced and should be independent from the incident beams size. Our experiment confirms this expectation. The absolute reflectivity is 65%, with an incident energy of $E_i = 28$ meV and a surface temperature of 90 K, using the smallest aperture of the rotatory diaphragm (400 μ m). Furthermore, as shown in Fig. 4, at $E_i = 28$ meV the intensity of the specular peak increases by a factor of 5 when we go from a diaphragm of 400 to one of 1200 μ m, while the FWHM remains constant. As far as we know, this is the highest value ever measured from a clean metal surface. These results are a clear indication of focusing of neutral He atoms by the thin Cu(111) crystal.

IV. CONCLUSIONS

In summary, four different thin crystals have been investigated: 100 μ m Cu(111), 50 and 100 μ m Ni(111), and 150 μ m Ru(0001). In order to select the surface that exhibits the best quality and the highest He reflectivity, an accurate analysis of their specular peaks has been carried out. The quality of the thin Ni(111) and Ru(0001) crystals was found to be worse than the corresponding bulk ones: the He reflectivity for the 100 μ m Ni(111) surface is 14%, decreasing to 3% for the 50 μ m one. Similar results have been obtained for the 150 μ m thin Ru(0001) crystal, whose absolute reflectivity is 5%. The corresponding average terrace size lies between 2–9 nm. In contrast, the quality of the 100 μ m Cu(111) crystal exceeds that of a bulk crystal, presenting both a high reflectivity (20%) and long-range crystalline order (>200 nm). A quite significant



FIG. 4. Comparison of He atoms scattered from Cu(111) surface by an incident beam with a collimation of 400 μ m at diaphragm position (black) and 1200 μ m (blue). The incident conditions are $E_i = 28$ meV and $\theta_i = 30^\circ$. The intensity of the specular peak increases by a factor of 5 while the FWHM remains constant when the diaphragm is changed from 400 to 1200 μ m.

- D. Farias and K. H. Rieder, Rep. Prog. Phys. 61, 1575 (1998).
- [2] M. Koch, S. Rehbein, G. Schmahl, T. Reisinger, G. Bracco, W. E. Ernst, and B. Holst, J. Microsc. 229, 1 (2008).
- [3] S. D. Eder, T. Reisinger, M. M. Greve, G. Bracco, and B. Holst, New J. Phys. 14, 073014 (2012).
- [4] S. D. Eder, X. Guo, T. Kaltenbacher, M. M. Greve, M. Kalläne, L. Kipp, and B. Holst, Phys. Rev. A 91, 043608 (2015).
- [5] B. Holst and W. Allison, Nature (London) **390**, 244 (1997).
- [6] K. Fladischer, H. Reingruber, T. Reisinger, V. Mayrhofer, W. E. Ernst, A. E. Ross, D. A. MacLaren, W. Allison, D. Litwin, J. Galas, S. Sitarek, P. Nieto, D. Barredo, D. Farías, R. Miranda, B. Surma, A. Miros, B. Piatkowski, E. Søndergard, and B. Holst, New J. Phys. **12**, 033018 (2010).
- [7] G. Comsa, Surf. Sci. 81, 57 (1979).
- [8] T. Reisinger and B. Holst, J. Vac. Sci. Technol. B 26, 2374 (2008).
- [9] B. Poelsema and G. Comsa, Springer Tracts in Modern Physics (Springer, Berlin, 1989), Vol. 115.
- [10] D. Barredo et al. Adv. Mater. 20, 3492 (2008).
- [11] A. Al Taleb, G. Anemone, D. Farías, and R. Miranda, Carbon 99, 416 (2016).

finding from the current study is that a beam of neutral He atoms has been focused to a spot of 350 μ m, with an absolute specular intensity two orders of magnitude larger than any He reflectivity previously reported for a flat or curved crystal. In order to improve the focal spot with this sample, a modification of the present experimental setup will be required. Finally, owing to their high quality, the thin Cu(111) crystals could be used instead of Cu metal foils when the crystallinity of the substrate is important to achieve best result, like for instance in the growth of large-area monocrystalline graphene [20] or hexagonal boron nitride [21].

ACKNOWLEDGMENTS

This work has been supported by the European Union, FP7: Theme NMP.2012.1.4–3 Grant No. 309672 and by the Spanish MINECO under project MAT2015–65356–C3–3–R (MINECO/FEDER). D.F. acknowledges financial support from the Spanish Ministry of Economy and Competitiveness, through the "María de Maeztu" Programme for Units of Excellence in R&D (MDM–2014–0377).

This work is dedicated to the memory of our mentor and colleague Karl-Heinz Rieder, pioneer in so many areas of surface science.

- [12] A. Politano, B. Borca, M. Minniti, J. J. Hinarejos, A. L. Vázquez de Parga, D. Farías, and R. Miranda, Phys. Rev. B 84, 035450 (2011).
- [13] P. Sutter, M. Minniti, P. Albrecht, D. Farías, R. Miranda, and E. Sutter, Appl. Phys. Lett. 99, 211907 (2011).
- [14] A. Tamtögl, E. Bahn, J. Zhu, P. Fouquet, J. Ellis, and W. Allison, J. Phys Chem. C 119, 25983 (2015).
- [15] A. Al Taleb, H. K. Yu, G. Anemone, D. Farías, and A. Wodtke, Carbon 95, 731 (2015)
- [16] H. K. Yu, K. Balasubramanian, K. Kim, J. L. Lee, M. Maiti, C. Ropers, J. Krieg, K. Kern, and A. M. Wodtke, ACS Nano 8, 8636 (2014).
- [17] P. Nieto, D. Barredo, D. Farías, and R. Miranda, J. Phys. Chem. A 115, 7283 (2011).
- [18] J. Lapujoulade, Y. Lejay, and G. Armand, Surf. Sci. 95, 107 (1980).
- [19] J. S. Becker, R. D. Brown, E. Johansson, N. S. Lewis, and S. J. Sibener, J. Chem. Phys. **133**, 104705 (2010).
- [20] V. L. Nguyen, B. G. Shin, D. L. Duong, S. T. Kim, D. Perello, Y. J. Lim, Q. H. Yuan, F. Ding, H. Y. Jeong, H. S. Shin, S. M. Lee, S. H. Chae, Q. A. Vu, S. H. Lee, and Y. H. Lee, Adv. Mater. 27, 1376 (2015).
- [21] H. Wang, X. Zhang, H. Liu, Z. Yin, J. Meng, J. Xia, X.-M. Meng, J. Wu, and J. You, Adv. Mater. 27, 8109 (2015).