High-resolution scattering apparatus for surface studies

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(Received 2 April 2002; accepted 1 September 2002)

A high-resolution apparatus designed to study the structural and dynamical surface properties is described. The apparatus combines the thermal energy He atom- and the low energy ion-scattering techniques both with time-of-flight detection of the scattered fraction. The energy spread of the supersonic He beam is less than 100 $\mu$eV at source temperatures below 35 K. The source temperature can be varied between 22 and 300 K to perform elastic, inelastic, and quasielastic scattering studies over a broad range of incident energies. The scattered beam is detected by a homemade quadrupole mass spectrometer designed to have high efficiency for He. The ion source works between 2 and 5 keV and the ion beam is mass selected by a Wien filter which also separates the transmitted ions from fast neutral species. The beam is detected at a scattering angle of 160° and therefore neutral impact collision ion scattering spectroscopy studies can be performed. The base pressure in the target chamber is in the 10$^{-11}$ mbar range. The capabilities of the apparatus are tested, and the evolution of surface disorder on Ag(110) up to 800 K is characterized. © 2002 American Institute of Physics. [DOI: 10.1063/1.1517147]

I. INTRODUCTION

Sophisticated and sensitive experimental techniques are required to investigate the structural and dynamical properties of surfaces. In fact, a detailed knowledge of these properties provides a deeper understanding of complex surface phenomena which are of fundamental interest for many applications, e.g., in nanotechnology to control atomic/molecular self-organization. Moreover, the knowledge has to be gained over a broad temperature range since the structure as well as the dynamics of the bare surface change with the temperature. This often leads to an evolution of the morphology or even to a loss of stability in nanostructured systems.$^{1,2}$ Theoretically and experimentally, it has been shown that as a result of increasing temperature surface defects proliferate, anharmonic vibrations and atomic diffusion are strongly enhanced, and the surface may roughen and melt ahead of the bulk.$^3$ To gain a whole insight into this large class of phenomena it is useful to investigate the local and the long range surface order simultaneously with high spatial and energetic resolution. To this aim, we have designed and constructed an experimental setup which combines the thermal energy atom scattering (TEAS) technique with low energy ion scattering spectroscopy (LEIS), both of them with time-of-flight (TOF) detection. These techniques are truly complementary since they provide reciprocal and real space information, respectively. TEAS is a well established diffraction technique strictly sensitive to the first surface layer. The most used probe is the helium beam with a wavelength of the order of 1 Å. He atoms are scattered by the tail of the surface electronic density a few angstroms above the topmost atomic cores. Therefore any perturbation or damage of the sample is excluded making TEAS a well suited technique to investigate surfaces covered with weakly bound or reactive adsorbed species.$^{4-6}$ Since the probe is neutral, TEAS also provides information on the structure of conductive long range ordered systems as well as insulating ones.$^{7-9}$ The introduction of the TOF detection makes TEAS also useful to investigate details of the surface lattice dynamics$^{10-15}$ and to study thermally disordered surfaces giving, for instance, information on diffusion, roughening, and melting mechanisms.$^{16-19}$

The LEIS technique is sensitive to the local properties of the surface because at beam energies above 1 keV the incoming ion wavelength is orders of magnitude smaller than the crystal lattice parameter. Hence the scattering process is not affected by coherence effects and can be classically described. With the introduction of TOF detection both charged and neutral scattered particles can be collected. This strongly enhances the sensitivity of the technique leading to a noticeable lowering of the ion dose used to perform measurements. Thanks to this improvement and because of the reduced penetration depth limited to a few atomic layers, LEIS is widely used to investigate the structure of clean and adsorbate covered metal and semiconductor surfaces. In particular, it provides information on nearest and next nearest neighbor distance, on the adsorption sites, and on the surface chemical composition.$^{20}$ The technique is also employed to study the evolution of the surface structure and dynamics with temperature. Indeed, the short range defect concentration and the lattice vibration amplitude can be measured up to the bulk melting point.$^{21-23}$

In this article we describe the scattering apparatus, and we report test measurements performed with both techniques on the (110) surface of silver in the temperature range between room temperature and 800 K. In particular, we point out how the complementary information provided by LEIS and TEAS gives a complete picture of the evolution of the
A. General description

The apparatus consists of three main different parts: the supersonic He source chamber (C1), the experimental chamber, and the detector chamber. There is a fourth small chamber which houses the He atom beam monitor (CM). The latter is particularly useful during the alignment procedure of the He atom beamline. Figure 1 displays a schematic top view of the apparatus. The section is made at the scattering plane level. We note that the experimental chamber is further divided into three different stages: C2, C3, and C4. The stage C3 is a box welded at the lower half of the experimental chamber. This provides more space at the upper level of the stage C4 which can be accessed by the sample. The usual surface cleaning-checking facilities [ion gun, Auger electron spectroscopy, and low-energy electron diffraction (LEED)] and the residual gas analyzer reside here. The detector chamber is divided in four stages: C5, C6, C7, and C8. The whole system can be separated into four sections by means of three gate valves positioned as shown in Fig. 2. In particular, the homemade valve between C2 and C3 has been specially designed with reduced thickness in order to be mounted inside the chamber. This modular design allows maintenance and repairing operations in a section without breaking the vacuum in the remaining sections of the apparatus. All the pumps connected to the different chambers are turbomolecular pumps except for the He source stage. In fact, C1 is pumped by a baffled diffusion pump bared by a blower and a rotary pump. The pumping speed in C1 can be further increased by ~30% with the turbomolecular pump T1. The rough ports of the turbomolecular pumps are generally connected to another turbomolecular pump to increase the compression ratio for He. This is useful especially for the detector chamber where a very low He partial pressure is required to obtain a satisfactory signal-to-noise ratio. The base pressures in stages C1–C8 with the He-beam on and off are reported in Table I along with the corresponding pumping speeds.

B. Sample manipulator

The sample S is located at the center of the experimental chamber in the stage C4. The molybdenum sample holder is mounted on a manipulator which allows three rotational and three translational degrees of freedom. The vertical translation moves the sample between the upper service space and the scattering plane level of the chamber. The two horizontal translations allow the precise alignment of the crystal surface at the intersection of the incident He and ion beams and the structural and dynamical properties of the outermost surface layers.

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The noble gas ion beam is produced and accelerated up to 5 keV by a differentially pumped commercial ion gun with kept stable within 0.02 K in the range 22–300 K by means of a temperature controller. A mount with two rotational degrees of freedom and three translations allows a careful alignment of the nozzle with respect to the skimmer as well as changes of the nozzle-skimmer distance, \( d_{ns} \). The pressure at the blower’s inlet is measured with a capacitance diaphragm gauge and this value is multiplied by the pumping speed of the blower itself to evaluate the nozzle He flow. The maximum flow at the roll off pressure of the diffusion pump is about 5 mbar l/s. The He beam which enters C2 is chopped in short pulses by a rotating slotted disk at \( \sim 20 \) cm from the skimmer tip. The disk is mounted on the axis of a synchronous motor driven by a controller whose frequency is stabilized within 0.01% by means of a quartz-controlled oscillator. The influence of frequency fluctuations on the measured time distribution of the beam is therefore negligible. When the synchronous motor rotation is phase locked with the controller, the motor frequency \( f \) can be varied in the range between 250 and 800 Hz. Two sequences of slits are machined on the periphery of the chopper disk at an angular distance of 180°. Each sequence consists of three slits corresponding to duty cycles of 5%, 2%, and 1%. The shortest opening time is \( \tau = 4.3 \) \( \mu \)s at \( f = 800 \) Hz. To select the optimal duty cycle, the motor is mounted on a translation stage driven by a feedthrough. A LED-photodiode assembly mounted on the translation stage generates the reference signal for the acquisition system. Afterwards, the He beam is collimated by two small holes at both ends of the stage C3 before hitting the sample. The scattered beam enters the detector chamber mounted at \( \theta_1 = 110° \) with respect to the source axis. Here, it goes through the differentially pumped stages C5 and C6, it is ionized in C7, and is mass selected in C8. The ionizer has been designed with two lenses which focus the electrons emitted from the hot filament within a small ionization region. This design is similar to that of Ref. 26. In fact, high-resolution measurements require a short ionization region because it provides a contribution \( \Delta t_p \times L \) to the time spread of the measured beam. Trajectory simulations within the present geometry suggest an ionization length of about 4 mm. The \( \text{He}^+ \) ions are mass selected by a homemade quadrupole mass filter analyzer designed to work on low masses (up to 20 amu) with a transmission greater than 95% at mass resolution \( \sim 10 \). To fulfill these requirements, quadrupole rods with 30 mm radius have been used for the construction of the mass filter. At the mass filter exit, a repeller electrode deflects the ions on the first dynode of an electron multiplier. The multiplier output signal is amplified, discriminated, and fed to a multichannel scaler. The channel width is set to 1 \( \mu \)s with a negligible dead time between the channels. After averaging the signal over as many as 10\(^6\) repetitions, the TOF distribution is stored in the personal computer that controls the sample temperature and the angular positions. Figure 4 displays the geometry of the beamline along with collimator diameters and distances.

C. He atom beamline

The supersonic He source assembly is shown in detail in Fig. 3. The nozzle is cooled down to \( \sim 20 \) K by a two stage cold head and heated up by two resistive heaters. The source temperature is read by a platinum thermometer and two detector axes. The crystal can be tilted by \( \pm 2° \) to set the surface normal in the scattering plane (tilt angle \( \chi \)), can be rotated by 360° around a vertical axis to change the incident angle (polar rotation \( \theta \)), and can be rotated by 110° around the surface normal to change the crystal direction within the scattering plane (azimuthal rotation \( \phi \)). The three rotations are driven by stepper motors controlled by a personal computer. The resolution and accuracy are a few hundredths of degree for \( \chi \) and \( \phi \) rotation while they are slightly better than 0.01° for the polar rotation. The crystal holder is linked to the bottom of a liquid nitrogen cryostat by means of a copper braid that is in contact with the cryostat through a sapphire disk. In this way the sample is electrically insulated from the apparatus and the ion current on the crystal can be measured. The sample holder is heated by electron bombardment and the sample temperature can be varied in the range 160–1000 K. The crystal temperature is measured by a K-type thermocouple. The temperature controller that drives the power supplied to the heating filament is interfaced to a personal computer.

D. Ion beamline

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The scattered particles sample is any misalignment of the beam. The average current on the sample while another couple of deflection plates corrects for and 6. Then, an Einzel lens focuses the pulsed beam on the mer. The appropriate pulsing sequence is described in Figs. 5 and 6. Then, an Einzel lens focuses the pulsed beam on the sample while another couple of deflection plates corrects for any misalignment of the beam. The current average on the sample is \( \sim 0.1 \text{nA} \) with a spot diameter of about 1.0 mm. The scattered particles (neutrons and ions) are detected in the backscattering geometry at an angle of 160° with respect to the source axis. The detector consists of two microchannel plates in a V-stack assembly set \( \sim 1 \text{m} \) far from the surface. The detector full acceptance angle is 0.6°. The signal is acquired with the same multichannel scaler used on the atom beamline but with a 200 ns channel width. The acquisition chain is also sketched in Fig. 5.

### III. CHARACTERIZATION OF THE BEAMLINES AND TEST OF THE APPARATUS

#### A. Performances of the supersonic He source

The He-atom beamline has been initially tested with the detector mounted in the position of the He beam monitor. In this configuration, the skimmer-ionizer distance is \( \sim 169 \text{cm} \). Time-of-flight spectra have been collected at different nozzle-skimmer distances to determine the best working condition for the He source. For each value of \( d_{ns} \), the source temperature, \( T_0 \), and the source absolute pressure, \( P_0 \), have been varied in the range 20–80 K and 2–16 bar, respectively. The spectra have been analyzed to estimate the parameters of the beam distribution, i.e., the most probable energy, the energy width, and the intensity of the He beam. This has been performed by fitting each spectrum with the sum of a constant, which represents the He background in the detector stage, and a function \( f \) describing the distribution of the He-atom beam. This function is the Maxwellian distribution converted in a time-of-flight scale, \( f(t) = A(t_0/t)^5 \exp\left(-[(t_0/t) - 1]^2/\sigma^2\right) \), where \( A \), \( t_0 \), and \( \sigma \) are the fitting parameters.  

The peak is integrated to estimate the beam intensity while the contributions of finite chopper opening time (\( \tau \)) and ionization time length (\( \Delta T_D \)) are subtracted from the measured full width at half maximum of the peak \( (\Delta T_{exp}) \) to obtain the intrinsic time spread of the beam \( (\Delta T_B) \):

$$\Delta T_B = \sqrt{\Delta T_{exp}^2 - \tau^2 - \Delta T_D^2}.$$  

\( \Delta T_B \) is finally used to calculate the incoming beam energy width \( \Delta E \). The results at \( d_{ns} = 20 \text{mm} \) are reported in panels (a) and (b) of Fig. 7. The highest intensity is obtained at source parameters \( P_0 = 13 \text{bar} \) and \( T_0 \sim 60 \text{K} \) corresponding to \( \Delta E \sim 200 \mu\text{eV} \). On the other hand, an energy width less than 100 \( \mu\text{eV} \) is achieved at temperatures below 35 K and

**FIG. 4.** Diagram of the atom beamline: the He path lengths and the diameters of the collimators are reported. All values are given in mm.

**FIG. 5.** The ion beamline is sketched together with the block diagram of the pulsing sequence and acquisition chain. The incoming ions are electrostatically deflected in the \( x-y \) plane perpendicular to the beam axis by two couples of plates connected with two pulse generators (Ref. 45). The \( V_{xp} \) pulse is delayed with respect to the \( V_{yp} \) pulse. During the pulsing sequence (see Fig. 6 for details) the beam sweeps over the skimmer entrance and an ion pulse having a time width between 50 and 100 ns is generated. On the rising edge of \( V_{yp} \), a trigger signal is sent to the multichannel scaler (MCS) which starts the time-of-flight data acquisition. Once completed, the measured spectrum is stored on the hard disk of a personal computer.

**FIG. 6.** Details of the pulsing sequence of the ion beamline. Static potentials on the X and Y plates deflect the incoming beam in A. The pulsing sequence drives the beam along the trajectory \( ABCD \). The beam sweeps over the skimmer entrance on the rising edge of \( V_{yp} \).
$P_0 \leq 4$ bar with a beam intensity still greater than $30\%$ of the maximum value. Moreover, panels (a) and (b) of Fig. 7 show that for each given $P_0$ there is a temperature where $\Delta E$ presents a minimum which approximately corresponds to a maximum of the intensity. In panel (c), the measured beam energy widths at $P_0 = 4$ and 6 bar are compared with the theoretical results obtained by using the moment method of Ref. 31 to solve the Boltzmann equation for a free jet expansion. Calculations are performed assuming an ellipsoidal velocity distribution function and an analytical He–He van der Waals potential based on perturbation theory. The influence of quantum effects on the flow properties is taken into account. Deviations from theory are observed whenever the calculated beam temperature along the source axis decreases below $\sim 10^{-3} \text{ K}$ during the expansion, that is at $T_0 \leq 40 \text{ K}$ for $P_0 = 4$ bar and $T_0 \leq 50 \text{ K}$ for $P_0 = 6$ bar. Measurements performed on the $\text{He}_2^+$ signal have shown that in these cases the dimer ion fraction is greater than $2 \times 10^{-4}$. This suggests that the corresponding cluster concentration together with residual gas scattering during the expansion contribute to the beam heating which is observed as an increase of the energy width.

The detector chamber has been finally mounted in the configuration shown in Fig. 1. Measurements performed on the $\{110\}$ surface of silver along the $\langle 110 \rangle$ azimuthal direction are shown in Fig. 8. The incoherent elastic peak measured at two sample temperatures is compared with the apparatus response function estimated with the sample held at room temperature. Above $600 \text{ K}$ a slight peak broadening of a few tenths of $\mu$eV is observed. This is due to small energy transfers between the He beam and the adsorbed silver atoms diffusing along the surface. Measurements of the so-called quasielastic peak broadening at different temperatures and parallel momentum transfers provide information on the surface diffusion process. In particular, an estimation of the surface diffusion coefficient $D = \langle 9.1 \pm 0.4 \rangle \times 10^{-6} \text{ cm}^2/\text{s}$ is obtained at $650 \text{ K}$ within the single jump diffusion model. The estimated value is in good agreement with the predictions of molecular dynamics simulations. Assuming that the evolution of $D$ with temperature is well described between 600 and $800 \text{ K}$ by the usual Arrhenius law, an estimation of the energy barrier for hopping yields a value of $0.29 \pm 0.02 \text{ eV}$. This is, at least to our knowledge, the first quasielastic He atom scattering study of surface self-diffusion in the intermediate temperature range, i.e., around half the bulk melting point ($T_{\text{mp}} = 1234 \text{ K}$ for Ag).

### B. Performances of the ion line

The ion beamline has been characterized on the same Ag(110) sample in the temperature range between 300 and $700 \text{ K}$. An azimuthal pattern (intensity vs $\phi$) measured with $2 \text{ keV} \text{ Ne}^+$ ions at grazing incidence and slightly above room temperature is displayed in Fig. 9. The deep minima are due to channeling effects along the main crystal directions and are typical for a well ordered fcc (110) surface structure shown in the inset. From the angular position of the minima, a given azimuthal direction can be set in the scattering plane within $\pm 1^\circ$, and the evolution of the scattered intensity vs the incidence angle is measured to obtain neutral impact collision ion scattering spectroscopy (NICISS) spectra. Figure 10 shows the NICISS scans measured along the $\langle 001 \rangle$ azimuth at $500 \text{ K}$ (inverted triangles) with $4 \text{ (a)}$ and $2 \text{ keV (b)} \text{ Ne}^+$ beams, respectively. Two focusing peaks (1 and 3) are well resolved. As sketched in the inset, the structure at incidence angle around $50^\circ$ can be mainly ascribed to particles focused by the first layer atoms on the third layer atoms. As a result of the beam energy...
increase, this feature gains intensity over that at lower incidence angle. We note that the structure (3) was scarcely visible in a previous experiment carried out at \( \sim 3 \) keV without neutral detection.\(^{40}\) In Fig. 10(b), the evolution with temperature of the NICISS intensity measured with a 2 keV Ne\(^+\) beam is reported. It can be noted that the first focusing peak broadens because of the enhanced lattice mean square vibration amplitude. Moreover, scattering contributions which can be ascribed to surface defects like vacancies and adsorbed atoms appear at grazing incidence angle.\(^{21}\) The increase of surface disorder also causes the third surface layer to become more visible. Information on the evolution with temperature of the mean square displacement of the top layer atoms is gained by fitting the first focusing peak within the two atom scattering model described in Ref. 23. In the inset, a section of the surface parallel to the scattering plane is reported showing the trajectories which mainly contribute to the 1 and 3 focusing peaks are also sketched.

**IV. DISCUSSION**

This article points out the peculiarities of a new experimental setup used to study structural and dynamical surface properties. LEIS and TEAS measurements performed on the (110) surface of silver show that up to 600 K lattice vibrations are harmonic and atomic diffusion is not detected. Above 600 K, LEIS results show an enhancement of the surface atom vibration amplitudes with respect to the prediction of the Debye model along with an increase of the surface defect density. This suggests that atoms of the topmost layer in their displacements from the equilibrium position are probing the anharmonic part of the potential and, with a finite probability, they can overcome the energy barrier for getting adsorbed onto the surface. Here they can diffuse causing the quasielastic broadening observed by He scattering. Estimations of the diffusion coefficient and the energy barrier for surface hopping in the temperature range between 600 and 800 K show a very good agreement with the calculated static energy barrier\(^{25}\) and with STM results obtained below room temperature.\(^{3,44}\)

**ACKNOWLEDGMENTS**

The authors gratefully acknowledge R. De Ferrari, A. Ferrando, A. Perelli, and A. Pizzorno for their useful contribution to the design and the setup of the apparatus. The authors are very grateful to G. Maloberti, E. Cavanna, and P. Pollovio for technical support. The work has been supported by the Italian MURST (Grants No. 9702178261-003 and No. 9902112831), by the Consiglio Nazionale delle Ricerche through the project Superfici ad Alta Temperatura, and by the Surfaces and Interfaces section (F) of the Istituto Nazionale di Fisica della Materia.

25 Autotuning Temperature Controller, Model 330, Lake Shore, Westerville, OH.
27 Multichannel Scaler, model Turbo MCS and Fast Preamplifier, model VT120, E.G. & G. Ortec, Oak Ridge, TN.
28 Ion Source, model IQE 12/38 and Wien Filter, model WF-IQE, SPECS GmbH, Berlin, Germany.
45 Universal Pulse Generator, model 6040, Berkley Nucleonic Corporation, CA.