THE STUDIES OF SURFACE LAYER ATOMIC STRUCTURE OF AL(100) BY MEV ION SCATTERING

Cheng Huansheng (承焕生), Cui Zhixiang(崔志祥), Xu Hongjie (徐洪杰),
Yao Xiaowei (要小未) and Yang Fujia (杨福家)

(Fudan University, Shanghai 200433, China)

(Received November 1989)

ABSTRACT

A UHV system specially designed for studying surface and interface atomic structure by MeV ion scattering and channeling is described. The vacuum in the UHV chamber is 133.332×10^{-10}Pa. The chamber is equipped with an ion gun used for sample cleaning, a translatable four－grid LEED－Auger system used for characterization of the crystal surface, and a three dimensional goniometer. The crystal preparation and cleaning procedure of Al(100) are presented. The surface peak intensity of Al(100)－<100> and Al(100)－<100> has been measured by MeV ion channeling and scattering. The measured surface peak intensity was compared with that of Monte－Carlo simulation. The experimental results indicate that the thermal vibration amplitude of Al(100) surface atoms is 1.2－1.3 times that of bulk atoms. The relaxation of first layer for Al(100) is less than －0.005nm.

Key words: MeV ion scattering Surface layer atomic structure Al

I. INTRODUCTION

Knowledge of the atomic configuration is essential to understanding and controlling electronic, chemical and mechanical processes at crystal surfaces and interfaces. During the past 10 years high energy ion scattering spectroscopy has emerged as a powerful and direct tool to get such information[1-2]. this technique is particularly useful for the study of atomic displacements with respect to regular lattice sites. It appears therefore to complementary to low－energy electron diffraction(LEED) measurements, which give information mainly related to crystallographic symmetries and for which rather extensive calculations and strong assumptions are required to evaluate the static (defect) or dynamic (thermal vibrations) atomic displacements. The major advantage of this technique is that a simple Monte－Carlo simulation can be performed and compared to experimental results. In this calculation a description of the crystal (mean atomic positions and thermal vibrations) is proposed, and the only physical value required to simulate the experiment in this crystal is the interatomic potential between the incoming ion and a crystal atom. Since this potential is known with high precision, a comparison between the simulations and experimental results is a good check on the crystal description assumed. In practice the crystal surface structure can be checked by comparison between simulation and experiment for various incident－beam directions, beam energy, crystal temperatures, etc. In this paper we report the experimental results of

surface atomic structure for Al(100) by MeV ion scattering and channeling.

II. EXPERIMENTAL

The experiments were performed in a UHV chamber, which was specially designed* * and built for study of surface and interface atomic structure by MeV ion scattering and channeling. Fig.1 shows the experimental apparatus. Ion and titanium sublimation pumps maintain the chamber at 133.332×10^{-10}Pa. The UHV chamber is coupled via a differentially pumped beamline to a 3 MV Tandem accelerator. The beamline, coupled to the chamber by a 1 mm diam. collimating aperture, is held at 133.332×10^{-8}Pa by a turbomolecular pump. At the upstream end of the beamline, another collimator provides further isolation from the accelerator and beam switching magnet, which are in the 133.332×10^{-7}Pa range.

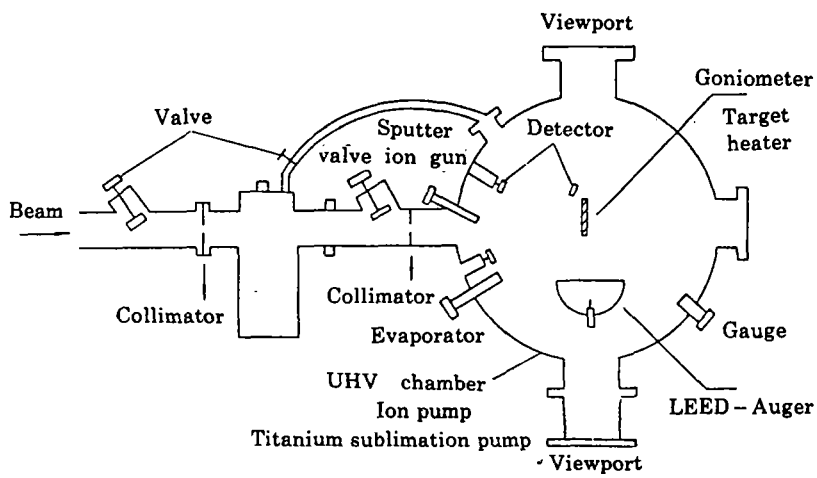

Fig.1 Schematic of the experimental apparatus described in the text.

The chamber is equipped with a sputtering ion gun for sample cleaning, and a horizontally translatable four – grid LEED – Auger system used for characterization of sample surfaces. The LEED – Auger system is provided with a transparent screen. This feature is required since the sample holder is large, and hence the observation of the LEED patterns is much easier and precise from the electron – gun side. A leak valve manifold permits the admission of variety of gases for sputtering or absorption studies, this relatively high pressure gas may then be removed rapidly by the turbopump through an auxiliary exhaust line which bypasses the chamber's beam entrance aperture.

The sample manipulator used in these experiments is based on a UHV concentric rotary and linear motion feedthrough mounted on a movable stage. It permits three independent translations, a tilt, and two rotations: one about an axis in the sample surface ($\pm 180^{\circ}$. in 5 range, 1′ graduations) and one about an axis normal to the sample (360°, 4′ resolution). The sample may be heated by electron bombardment from the back side, and its temperature is monitored with a thermocouple, which was checked by a optical pyrometer.

The energy of scattering ions is measured with a special bakeable semiconductor detector from Ortec. The grazing exit angle geometry is used to improve the depth resolution and reduce the background under the surface peak. Many ports on the chamber can be used for viewing and for the installation of further equipment such as an evaporator and a quadrupole mass spectrometer.

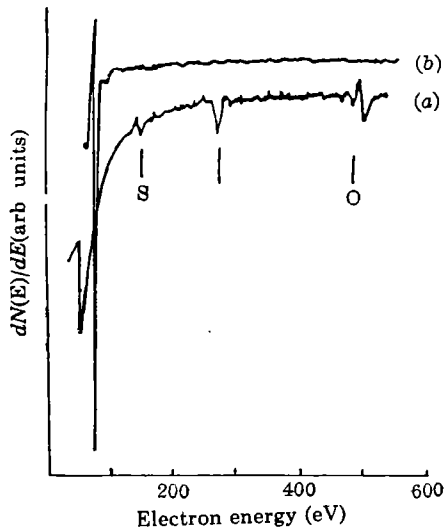

Fig.2 Auger spectrum of Al(100)

(*a*) Before cleaning procedure

(*b*) 10 h of Ar ion bombardment + annealing.

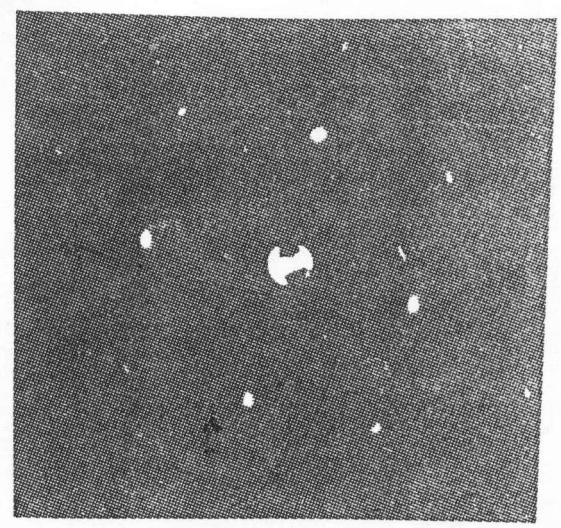

Fig.3 LEED pattern of Al(100) (1×1) at 78eV

The Al(100) single crystal sample, 10×20×1.5 mm³, was cut from a Al bar of high purity. The slice was first mechanically polished to within 0.5° of the desired crystal plane using an X－ray goniometer, then electropolished in a solution of perchloric acid (with density of 1.67 g/ml), ethanol and deionized water in a volumetric ratio of 3:4:7. This operation removed the impurities and a 2μ m thick damaged layer which were introduced by mechanical polishing, and resulted in bright mirror－like surface. The sample cleaning procedure in the UHV chamber consists of a cyclic treatment of Ar－ion bombardments (10μ A, 500V,1H) and annealing (700K, 0.5H). The Ar beam hits the surface under 45° incidence. During argon bombardment, argon is introduced into the chamber at a 666.66×10^{-5}Pa pressure, and the ionic pumping is stopped. In order to lower the contaminant (CO,H_2O) partial pressure, the titanium sublimation is performed before argon introduction. The cleaning process was repeated until only Al signal and no trace of impurities could be seen from Auger spectra. Before cleaning treatments, the C,O(mainly from native Al O film on the surface)and S were present. When these impurities were removed, the Auger signals from Al increased rapidly and the main Al peak moved to 68 eV from 55 eV (which is from Alumina). Fig.2 shows the Auger spectra from Al. The LEED patterns following these cleaning treatments showed a sharp (1×1) structure. Fig.3 shows the LEED pattern of Al(100) (1×1).

Ⅲ. MONTE CARLO SIMULATIONS

The experimental results were compared to Monte－Carlo simulations. A

Monte-Carlo computer simulation, incorporating the binary collision approximation for an atomic row, was used to perform the calculations. The ion-atom interaction was modeled by means of the Moliere expression[3] for the screened coulomb potential with a fully ionized screening parameter appropriate to light ions in the MeV energy range. Evaluation of the surface peak intensity from the atomic row, in atoms/row, was done by the nuclear encounter probability (NEP) technique of Barrett[4]. In all the calculations, the number of atom in the rows, the number of ion trajectories and the number of independant runs were large enough to ensure a high degree of statistical precision in the results.

The thermal vibration-induced displacement of the target atoms from the equilibrium position was determined randomly at the instant of collision under the assumption of a Gaussian distribution

$$P(x) = [1/(2u_1^2)^{1/2}] \exp(-X^2/2u_1^2) \quad (1)$$

where u_1 is determined by the Debye theory provided the Debye temperature and the target temperature are given. The Debye temperature of Al is 428K[5]. If we assumed the target temperature to be 293K, then the thermal vibration amplitude u_1 is calculated to be 0.0096nm by the Debye approximation. Correlations of thermal vibrations were introduced through the nearest-neighbor correlation coefficient, C, defined by

$$C = <\triangle X_m \triangle X_{m+1}>/<\triangle X_m^2> \quad (2)$$

where $\triangle X_m$ is the displacement of the mth atom from the normal site and $<>$ indicates the time average. The value of C ranges from 0.14-0.36 at low and high temperatures[6]. Using the model described in Ref. [6], C = 0.2 and 0.3 were calculated for Al<100> and Al<110> area, respectively.

The vibration amplitudes of surface atoms are significantly greater than those of atoms in the bulk of the solid due to the asymmetric forces experienced by the atoms at the surface. This enhanced thermal vibration was estimated assuming that the thermal vibration amplitude of atoms at the first monolayer is $\varepsilon\ u_1(\varepsilon \geqslant 1)$ and that the enhancement decays into the bulk exponentially with a decay constant of 0.405nm (the lattice constant of the Al crystal [7]). The value of the enhancement factor for Al(100) is unknown. In this study ε is obtained from the best fit of our experimental results to Monte-Carlo simulation.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

Fig.4 shows the <100> axial channeling dip from the Al(100) sample treated by the above mentioned cleaning procedure. The energy of incident helium ions was 1.2 MeV. A $\psi_{1/2}$ of 0.55°, which is in good agreement with expected from calculation, and η_{min} of 3.5% behind the surface were measured. Considering the Debye temperature of 428 K for Al, a η_{min} of 4.0% is predicted from calculation. It confirms the presence of good crystal order in subsurface crystal planes. A typical channeling spectrum obtained using a 1.0 MeV He^+ beam incident along the normal<100> axis is given in Fig.5.

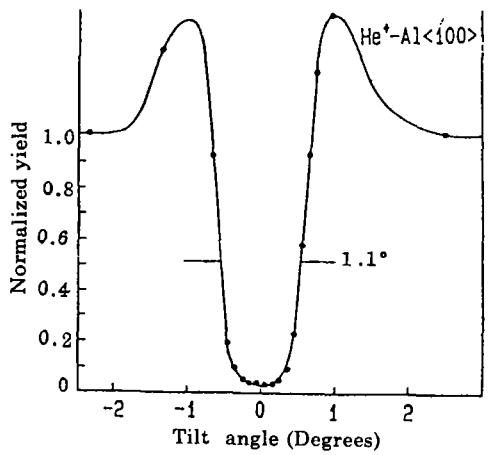

Fig.4 Al ⟨100⟩ axis channeling dip after cleaning

Incident ion energy E_{He^+} = 1.2MeV

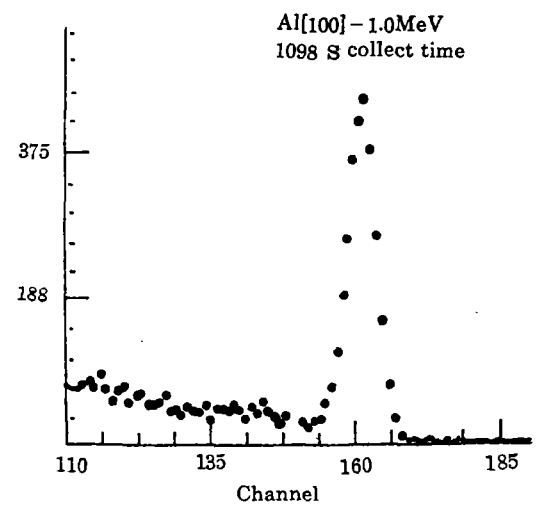

Fig.5 Typical energy spectrum from clean Al(100)with the beam in the surface normal ⟨100⟩ direction and the detector at ascattering angle of 95°

The spectrum is taken with 3UC of 1.0 MeV He^+ incident.

To determine the surface peak yield, a background subtraction was performed using the triangular correction. The background contribution from bulk is about 3% of surface peak area for incident ion energy of 1.0 MeV; it slightly increases with decreasing incident ion energy and reaches 6% of surface peak area at 0.6 MeV. In order to determine the significance of beam dose effects, the surface peak yield and subsurface scattering yield were measured as a function of dose. For 1.0 MeV He^+ ions incident along the Al<100> direction neither the change in surface peak yield nor in sub-surface scattering yield was detected for doses up to 100 μ c/mm² (implanted He^+ ions 6×10^{18} ions/cm²). Most measurements in this work were recorded with ion doses below 50 μ c/mm² for a beam spot. We conclude that the measured surface peak yields are not influenced by beam damage. The measured surface peak yields were converted into atoms/cm² or the more convenient unit atoms/row using a standard, which is a silicon sample covered a thin Ta film with known area density[8]. The absolute surface peak yields reported here should be accurate to within 4%.

Fig.6 shows the energy dependence of the surface peak intensity for clean Al(100) in the normal <100> direction. Energy of incident ions was varied from 0.6- 2.0 MeV. The solid line is the result of computer simulations assuming a bulk-like structure and thermal vibration amplitude (u_1 = 0.0096nm), a vibration correlation coefficient (c = 0.2), and an enhanced surface thermal vibration (enhanced factor ε = 1.2 for the first layer, 1.1 for the second, and 1.0 for the third and deep layers). The measured

surface peak intensity is in good agreement with the prediction of the calculation. The dashed line in Fig.6 is what is predicted by computer simulations for uncorrelated vibrations (u_1 = 0.0096nm, c = 0, ε =1.0). All of the surface peak yields (dashed line) are higher than that of measured results by ~5%. We note that the correlation effects are the only known mechanism which results in a decrease of the surface peak intensity from the uncorrelated values. All other conceivable phenomena, such as surface reconstructions, enhanced surface vibrations or radiation damage in the surface act to increase the surface peak intensity. The good agreement between calculation and experiment confirms the effect of vibration correlations.

Fig.7 shows the energy dependence of the surface peak intensity for Al(100) in the off <110> normal direction. The solid curve is the prediction of computer simulations assuming u of 0.0096nm, C of 0.3 and an enhanced surface vibration (ε = 1.3 for the first layer, 1.15 for the second and +1.0 for the third and deeper layer). The enhanced surface vibration can be described with effective surface Debye temperature[9] Q (normal to the surface) and Q (parallel to the surface). The Q of 354K resulted from the surface peak intensity for Al(100) in <100> direction. The measured surface peak intensity for Al(100) in the <100> direction depends on the vibration amplitude of surface atoms, u<100>, perpendicular to that row, and u<100> has contributions from u together with u of the Al(100) surface.

$$u^2 <110> = u \cos^2\alpha + u^2 \sin^2\alpha \qquad (3)$$

where α is the angle between <100> and <110> of Al. The value of α is 45°. A value of 300K for Q is obtained for the Al(100) surface. Jackson reported[9] Q and Q are 361K and 223K, respectively from calculation using a decouple osillator model. The calculated Q is in good agreement with that of our experiment; but the value for Q is 26% lower than that of our measurement. The vibration amplitude of surface atoms can also be determined by LEED or Mossbauer effects, but so far there is no other report of experimental results on Al(100) surface vibration.

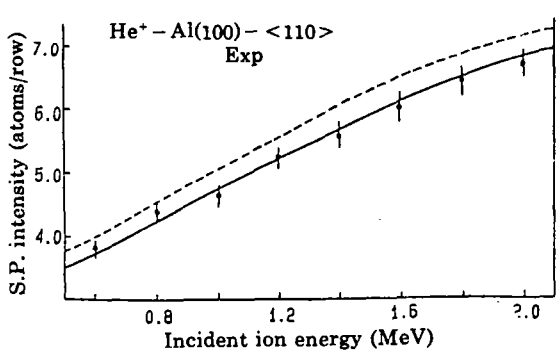

Fig.6 Energy dependence of surface peak intensity for Al(100)- <100>

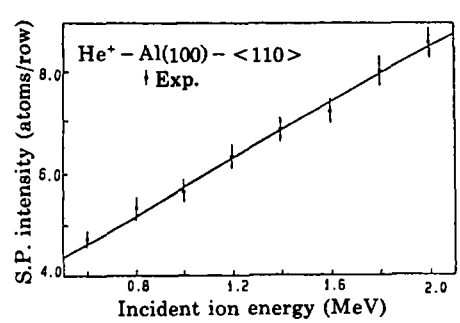

Fig.7 energy dependence of surface peak intensity for Al(100) in the off normal <110> direction

Fig.8 shows the angular dependence of surface peak intensity near the <110> axis measured along the (100) plane normal to the surface. The energy of incident ions was 1.0 MeV. Solid and dashed curves are from Monte-Carlo simulations in which a bulk-like structure with enhanced surface vibration and contraction (relaxation inward) of the first layer by 0.005nm. The center position of the solid curve is at 45° with respect to the <100> axis, which was determined by minimum yields of the <110> axis bulk dip. If no surface relaxation occurs on this surface, the angular distribution of the surface peak intensity should be symmetric about the bulk <110> direction. For relaxation inward (contraction) there should be an asymmetry towards larger angles and for relaxation outward (expansion) towards smaller angles relative to the bulk <110> direction. The measured results are slightly shifted and in

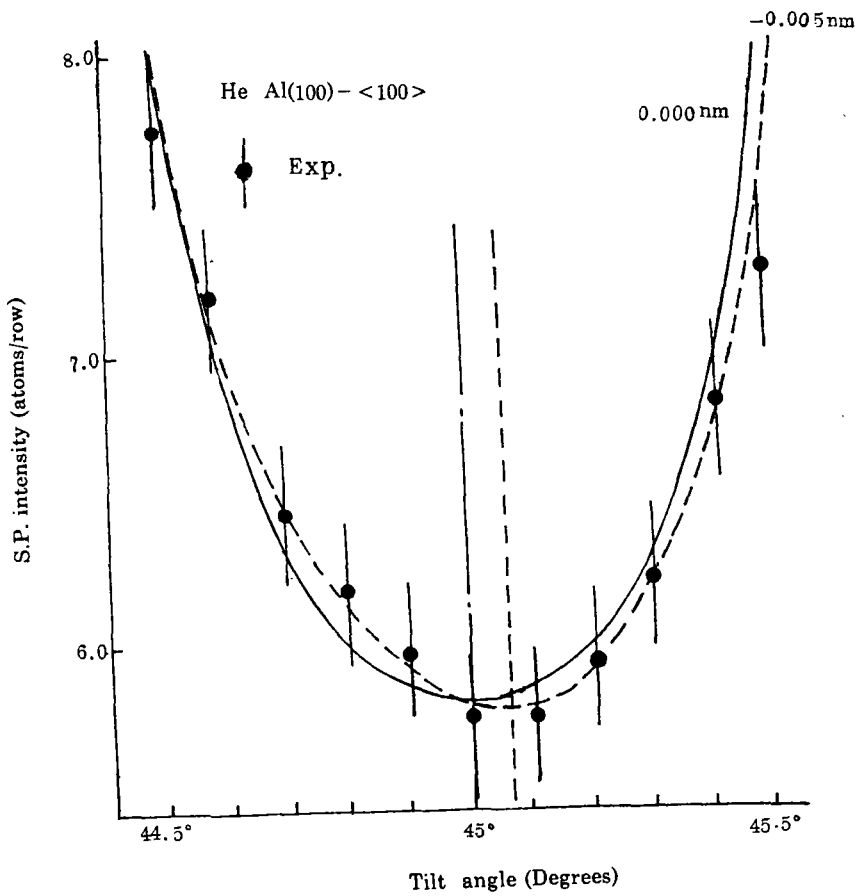

Fig.8 Angular dependence of surface peak intensity near the <110> axis

E_{He} = 1.0 MeV. Solid and dashed curves are from Monte-Carlo simulations in which a bulk-like structure with enhanced surface vibration and contraction of the first layer by 0.005 nm

agreement with the dashed curve. Considering the experimental error, it can be concluded that contraction of first layer for Al(100) is less than 0.005nm. This result is in agreement with the LEED studies[10], which found the first interlayer spacing to be equal to that of bulk within approximately 0.01nm. This result is also in agreement with that of from MEED[11] and EXAFS[12] measurements. This experimental result is also consistent in the recent model calculations of surface structure reported by Landman et al[13].

IV. CONCLUSIONS

The experimental results of MeV ion scattering on Al(100) confirm the effect of vibration correlations and indicate the thermal vibration amplitude of the first layer for Al(100) is 1.2−1.3 times of that bulk atoms. The relaxation of first layer is less than −0.005nm. This result confirms the LEED studies in a straightforward and somewhat more quantitative manner.

ACKNOWLEDGEMENTS

It is a pleasure to acknowledge Prof. W.M.Gibson and Dr. Shin Hashimoto for their contribution to the UHV system. Authors also would like to thank Prof. Xie Xide for her support and useful advice.

This work is supported by the National Natural Science Foundation of China.

**UHV chamber is a collaborative research program between Fudan University and the Department of Physics at SUNY/Albany on atomic structures of surfaces and interfaces.

REFERENCES

[1] T.Narusawa et al., *Phys. Rev. Lett.*, 47 (1981), 417.

[2] L.C.Feldman et al., Material analysis by ion channeling, Academic Press, New York, 1982.

[3] G.Mcliere, *Z. Naturforsch.*, A2 (1947), 133.

[4] J.H.Barrett, *Phys. Rev.*, B3 (1971), 1527.

[5] Dwight E.Gray, American institute of physics handbook, 2nd edition , 1963.

[6] R.S.Nelson et al., *Phil. Mag.*, 7 (1962), 1385.

[7] I.Stensgard et al., *Surface Sci.*, 77 (1978), 513.

[8] S.Rigo et al., *Nucl. Instr. Meth.*, 149 (1978), 721.

[9] D.P.Jackson, *Surface Sci.*, 43 (1974), 431.

[10] F.Jona et al., *J. Phy. C: Solid State Phys.*, 13 (1980), L155.

[11] N.Masud et al., *Surface Sci.*, 133 (1983), 580.

[12] A.Bianconi and R.Z.Bachrach, *Phys. Rev. Lett.*, 42 (1979), 104.

[13] U.Landman et al., *Phys. Rev.*, B21 (1980), 448.