High-Resolution Angle-Resolved Photoemission Study of the Al(100) Single Crystal

J. Jiang,† M. Higashiguchi, N. Tobida, K. Tanaka, S. Fukuda, and H. Hayashi
Graduate School of Science, Hiroshima University,
Kagamiyama 1-3-1, Higashi-Hiroshima 739-8526, Japan
K. Shimada, H. Namatame, and M. Taniguchi
Hiroshima Synchrotron Radiation Center, Hiroshima University,
Kagamiyama 2-3-1, Higashi-Hiroshima 739-0046, Japan
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The free-electron-like surface-derived electronic state in Al(100) has been examined in detail by high-resolution angle-resolved photoemission spectroscopy (ARPES). We observed a kink structure in the energy band-dispersion at the energy of ~−40 meV, which is derived from the electron-phonon interaction. Based on the quantitative analyses of the ARPES line shapes, we have obtained the imaginary and real parts of the self-energy. The electron-phonon coupling parameter has been evaluated to be \( \lambda_{\text{el-ph}} = 0.67 \), which is much larger than the electron-electron coupling parameter \( \lambda_{\text{el-el}} = 0.06 \).

Keywords: Synchrotron radiation photoelectron spectroscopy; Aluminum; High index single crystal surfaces; Metallic surfaces; Phonons

I. INTRODUCTION

Recently, energy and momentum resolutions of angle-resolved photoemission spectroscopy (ARPES) have been improved drastically. Now one can examine fine electronic structures of solids that are directly related to the physical properties of solids. Using photon energies in the vacuum ultraviolet and soft X-ray region, ARPES spectra are sensitive to the electronic states at surface. Based on the quantitative analyses of high-resolution ARPES spectral features, one can evaluate magnitudes of the electron-phonon and electron-electron interactions at any points on the Fermi surfaces.

Aluminum is the textbook example of a trivalent nearly-free electron metal and a weak coupling superconductor. It has been well known that there exists a surface-derived state centered at the \( \bar{\Gamma} \) point of the surface Brillouin zone (SBZ) in the (100) surface [1–4]. However, there has been no report on the Fermi surface mapping or a detailed examination of the many-body interactions on the quasi-particles so far. We believe detailed quantitative examinations of the surface state in Al(100) will contribute to the fundamental understanding of the two-dimensional Fermi liquid.

In this study, we have done high-resolution ARPES of Al(100) to clarify the surface-derived Fermi surface, and to evaluate magnitudes of the many-body interactions on the quasi-particles near the Fermi level \( (E_F) \). We have found a kink structure in the energy band dispersion, and evaluated the coupling parameters of the electron-phonon and electron-electron interactions.

II. EXPERIMENTAL

ARPES experiments were performed on the linear undulator beamline (BL-1) of a compact electron-storage ring (HiSOR) at Hiroshima University [6]. High-resolution ARPES measurements were carried out using an angular mode of the hemispherical electron-energy analyzer (ESCA200, GAMMADATA-SCIENTA). We set the total energy resolution at \( \Delta E = 15 \) meV for high-resolution measurements at \( h\nu = 43 \) eV, and 150 meV for the Fermi surface mapping at \( h\nu = 163 \) and 167 eV. The angular resolution was \( \Delta \theta = 0.3^\circ \), yielding momentum resolution of \( \Delta k_{\parallel} = 0.017 \) \( \AA^{-1} \) for \( h\nu = 43 \) eV and \( \Delta k_{\parallel} = 0.034 \) \( \AA^{-1} \) for \( h\nu = 167 \) eV. The clean surface of Al(100) single crystal (purity 99.9999%) was obtained by repeated cycles of Ar\(^+\) sputtering (5 keV) over 10 hours to remove oxide layers at surface, and annealing at 400°C for 30 minutes to minimize surface roughness introduced by Ar\(^+\) sputtering. The amount of impurities such as C, O, and S on the surface was measured by detecting the Auger electron spectra. The sample was mounted on the liquid-He-flow-type 5-axis goniometer (i-GONIO LT, R-DEC Co.). By changing tilt and polar angles of the goniometer, we could perform the two-dimensional Fermi surface mapping in \( k \)-space. The sample temperature was set at \( T = 30 \) K for high-resolution measurements, and \( T = 300 \) K for the Fermi surface mapping. The pressure of the main chamber was \( 1 \times 10^{-8} \) Pa.

III. RESULTS AND DISCUSSIONS

Figure 1 exhibits Fermi surface mapping taken at \( h\nu = 163 \) eV and at \( T = 300 \) K. The dashed blue line indicates the boundary of the second SBZ. One can clearly observe circular surface-derived Fermi surfaces centered at the \( \bar{\Gamma} \) point of the second SBZ \( (k_{\parallel x}, k_{\parallel y}) = (1.55 \) \( \AA^{-1}, 1.55 \) \( \AA^{-1} \)\). The evaluated peak positions are indicated...
FIG. 1: ARPES Fermi surface mapping of Al(100) surface at $h\nu = 163$ eV and $T = 300$ K.

FIG. 2: ARPES result of Al(100) surface state band dispersion. $k_\parallel$ is relative to the $\Gamma$ point in the second SBZ (along $\Gamma$–M direction in Fig. 1).

by dots in Fig. 1. The surface-derived Fermi surface in the first SBZ cannot be clearly observed at this photon energy probably due to weak matrix element [1].

Figure 2 exhibits the energy-band dispersion of the Al(100) surface state along the $\Gamma$–M direction taken at $h\nu = 167$ eV and 300 K. One can clearly see a free-electron-like energy-band dispersion. By fitting with a parabolic function $\varepsilon_k = -\omega_0 + (\omega_0/k_F^2)k_\parallel^2$ (blue dashed line in Fig. 2), we determined the Fermi energy ($\omega_0$) and Fermi wave vector ($k_F$). We have determined the Fermi energy of the surface state as $\omega_0 = 2.63$ eV, which coincides well with the calculated value of 2.62 eV [3]. The Fermi wave vector was evaluated to be $k_F = 0.94 \pm 0.005 \, \text{Å}^{-1}$. By using the area of the Fermi surface $S_F = \pi k_F^2 = 2.78 \, \text{Å}^{-2}$, the carrier density ($n$) of the surface state is calculated to be $n = 2S_F/(2\pi)^2 = 1.4 \times 10^{15} \, \text{cm}^{-2}$.

The Fermi wave vector $k_F = 0.94 \, \text{Å}^{-1}$ was also obtained from the radius of the circular Fermi surface at $h\nu = 163$ eV in Fig. 1. We have changed photon energy from $h\nu = 163$ eV up to 185 eV with a step of $\Delta h\nu = 2$ eV, and confirmed that the size of the Fermi wave vector ($k_F$) and the Fermi energy ($\omega_0$) of the surface-derived state are independent of incident photon energy. It is a direct evidence that the surface state is localized at surface.

On the basis of the formula $m^* = \hbar^2[(d^2\varepsilon(k)/dk^2)]^{-1}$, the effective electron mass is evaluated as $m^* = 1.27 m_e$, here $m_e$ stands for the free-electron mass. The value of $m^*$ obtained in the present study is larger than that obtained previously $m^* = 1.18 m_e$ [1]. As shown below, the effective mass is further enhanced due to the electron-phonon interaction near $E_F$.

Figure 3(a) shows a high-resolution ARPES image plot of the surface-derived state near the Fermi level taken at $h\nu = 43$ eV and at $T = 30$ K. It has a sharp spectral feature, which is suitable for the detailed line shape analyses to elucidate many-body interactions. In order to quantitatively analyze the spectral shape, we used momentum distribution curves (MDCs). We have fitted a MDC with a Lorentzian on a linear background, and obtained the peak position and linewidth ($\delta k$). The blue points in Fig. 3(a) indicate thus evaluated peak positions.

Figure 3(c) shows the area surrounded by the blue square in Fig. 3(a). One can clearly recognize a kink structure at $\sim -40$ meV below $E_F$. Since the magnitude of the energy of the kink coincides well with the energy scale of the bulk Debye temperature of Al ($\Theta_D = 426$ K, $k_B\Theta_D = 37$ meV), it is reasonable to assume that the structure is derived from the electron-phonon interaction [7, 8].
In addition, there is another evidence for the origin of the kink structure if we look at the energy distribution curves (EDCs) near \( E_F \). Figure 3(b) shows three EDCs (1, 2, 3) obtained from cuts along broken lines in Fig. 3(a). LaShell and Jensen studied the Be(0001) surface state and determined the electron self-energy [5].

They indicated that the electron-phonon interaction produces a hump structure to the main peak in the ARPES spectra. Figure 3(b) shows that the spectral width becomes narrower as the peak approaches \( E_F \), and there appears a hump structure besides the main peak near \( E_F \). Observed spectral features are similar to those for Be(0001) surface state, indicating that the kink structure is derived from the electron-phonon interaction.

Next we examine the strength of many-body interactions in quantitative way. The ARPES spectral features are given by the single-particle spectral function \( A(k, \omega) \), which is related to the imaginary part of the single-particle Green’s function, \( G(k, \omega) \):

\[
A(k, \omega) = -\frac{1}{\pi} G(k, \omega) = -\frac{1}{\pi} \frac{\text{Im} \Sigma(k, \omega)}{\omega - \epsilon_0^k - \text{Re} \Sigma(k, \omega) + i\text{Im} \Sigma(k, \omega)^2},
\]

where \( \epsilon_0^k \) represents the energy of non-interacting band. In the present analyses, we have assumed that \( \epsilon_0^k \) is linear near \( E_F \) as shown by a solid line in Fig. 3(c). \( \text{Re} \Sigma(k, \omega) \) and \( \text{Im} \Sigma(k, \omega) \) are the real and imaginary parts of the self-energy (\( \Sigma \)) where direct information on the many-body interactions are included. \( \text{Re} \Sigma \) gives an energy shift from non-interaction band. \( \text{Im} \Sigma \) is related to the spectral width, \( \Gamma = 2|\text{Im} \Sigma(k, \omega)| \) [7, 8]. In the present analyses, we used the MDC width (\( \delta k \)) to estimate the \( \text{Im} \Sigma(k, \omega) \), namely, \( 2|\text{Im} \Sigma(k, \omega)| = \delta E = (d\delta E/dk)\delta k \), where \( d\delta E/dk \) is the gradient of the energy band.

In the case that electron-scattering processes such as the electron-phonon, electron-electron, and electron-impurity interactions can be regarded as independent, the lifetime broadening (\( \Gamma = 2|\text{Im} \Sigma(k, \omega)| \)) of the quasiparticles is given by the sum of each contribution: \( \Gamma = \Gamma_{\text{el-ph}} + \Gamma_{\text{el-el}} + \Gamma_{\text{el-imp}} \).

Figures 4(a) and 4(b) respectively show obtained \( \Gamma = 2|\text{Im} \Sigma(k, \omega)| \) and \( \text{Re} \Sigma(k, \omega) \). Solid and dashed lines in Fig. 4(a) exhibit theoretical \( \Gamma_{\text{el-ph}} \) and \( \Gamma_{\text{el-el}} \), respectively. Recently, Sklyadneva et al. have reported an \textit{ab initio} study of the electron-phonon interaction in the surface electronic states of Al(100), on the basis of the density-functional theory using a linear response approach in the plane-wave pseudopotential representation [4]. In the calculation of \( \Gamma_{\text{el-ph}} \), we used theoretical Eliashberg function (\( \alpha^2 F \)) [4].

One can see experimental \( \Gamma = 2|\text{Im} \Sigma(k, \omega)| \) is explained well by the theoretical curves \( \Gamma_{\text{el-ph}} \) and \( \Gamma_{\text{el-el}} \). Solid line in Fig. 4(b) indicates the theoretical \( \text{Re} \Sigma_{\text{el-ph}} \), which also agrees well with the experimental one. These results confirm that the kink structure originates from the electron-phonon interaction.

One can evaluate a dimensionless coupling parameter of the electron-phonon interaction as follows.

\[
\lambda_{\text{el-ph}} = -\frac{\partial \text{Re} \Sigma_{\text{el-ph}}}{\partial \omega} \bigg|_{\omega=0}. \tag{2}
\]

FIG. 4: Imaginary part (a) and real part (b) of self-energy of the surface-derived state in Al(100).

Using the observed gradient of \( \text{Re} \Sigma_{\text{el-ph}} \) at \( E_F \) (dashed line in Fig. 4(b)), the parameter is evaluated to be \( \lambda_{\text{el-ph}} = 0.67 \). We should note that the electron-phonon coupling parameter of the Al surface state is comparable with that for Be, \( \lambda = 0.7 \) [5].

The calculated electron-phonon coupling parameter is \( \lambda_{\text{el-ph}} = 0.51 \) at the \( \Gamma \) point (at \( \omega_0 = 2.62 \text{ eV} \)), and increases with energy [4]. Although the coupling parameter at \( E_F \) was not provided, calculated \( \text{Re} \Sigma_{\text{el-ph}} \) (solid line in Fig. 4(b)) using theoretical \( \alpha^2 F \) coincides well with the experimental one near \( E_F \). It clearly indicates experimental coupling parameter is in good agreement with the theoretical value.

In order to evaluate the magnitude of the electron-electron interaction, we fit the experimental \( 2|\text{Im} \Sigma| \) with \( 2|\lambda_\omega\omega^2[1/4 + \ln 2 - (1/2) \ln |\psi_n|]\omega_0^2 + C \) [9] for the energy region of \( -800 \text{ meV} \leq \omega \leq -50 \text{ meV} \). Here \( C \) is a constant, and the Fermi energy was set at \( \omega_0 = 2.63 \text{ eV} \). We have obtained \( 2\beta = 0.03 \text{ eV}^{-1} \). Using the Kramers-Kronig relation, we have evaluated the coupling parameter of the electron-electron interaction as \( \lambda_{\text{el-el}} = 4\beta\omega_0(1/2 + \ln 2)/\pi = 0.06 \) [10]. Near the Fermi level, the effective mass is further enhanced due to the electron-electron interaction, and becomes \( m^* = (1 + \lambda_{\text{el-ph}}) \times 1.27m_e = 2.1m_e \).

IV. CONCLUSION

We have performed a high-resolution ARPES study of the surface-derived state in Al(100). We have observed a circular surface-derived Fermi surface centered at the \( \Gamma \) point. The area of the Fermi surface (\( S_\text{F} = 2.78 \text{ Å}^2 \)), the Fermi energy (\( \omega_0 = 2.63 \text{ eV} \)), the Fermi wave vector (\( k_\text{F} = 0.94 \text{ Å}^{-1} \)) and the carrier density (\( n = 1.4 \times 10^{15} \text{ cm}^{-2} \))
have been determined. We have observed a clear kink structure in the energy-band dispersion at $\sim -40$ meV, which corresponds well to the energy scale of the bulk Debye temperature $k_B \Theta_D = 37$ meV. We have evaluated the electron-phonon and electron-electron coupling parameters as $\lambda_{el-ph} = 0.67$ and $\lambda_{el-el} = 0.06$, respectively. We have found that $\lambda_{el-ph}$ is much stronger than $\lambda_{el-el}$.

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[10] In the present analyses, we have assumed the imaginary part of the self-energy as

$$\text{Im} \Sigma_{el-el}(\omega) = -\beta \omega^2 \left[ \frac{1}{2} + \ln 2 - \frac{1}{2} \ln \left( \frac{\omega}{\omega_0} \right) \right] \quad \text{for} \quad |\omega| \leq \omega_0$$

and

$$\text{Im} \Sigma_{el-el}(\omega) = -\beta \omega^2 \left[ \frac{1}{4} + \ln 2 \right] \quad \text{for} \quad |\omega| > \omega_0.$$ Here $\omega_0$ is the Fermi energy. On the basis of the Kramers-Kronig relation, one can calculate the real part of the self-energy as follows.

$$\text{Re} \Sigma_{el-el}(\omega) = \frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{\text{Im} \Sigma_{el-el}(\omega')}{\omega' - \omega} d\omega'.$$

The coupling parameter can be calculated as follows.

$$\lambda_{el-el} = -\frac{\partial \text{Re} \Sigma_{el-el}(\omega)}{\partial \omega} \bigg|_{\omega=0} = -\frac{1}{\pi} \int_{-\infty}^{+\infty} \frac{\text{Im} \Sigma_{el-el}(\omega')}{\omega^2} d\omega'$$

$$= \left. \frac{4\beta \omega_0}{\pi} \left[ \frac{1}{2} + \ln 2 \right] \right|_{\omega=0} \sim \frac{4\beta \omega_0}{\pi} \times 1.193.$$