Decay of Shockley surface state by randomly adsorbed Bi atoms at Ag(111) surfaces

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The decay of the Shockley surface state by randomly adsorbed Bi atoms at Ag(111) surfaces was investigated. As the Bi coverage (θ_{Bi}) increased, the energy dispersion $E(k)$ was observed to deviate from the intrinsic downward parabolic dispersion of the Ag(111) Shockley surface state at the low-energy side in scanning tunneling microscopy dI/dV images. Meanwhile, scanning tunneling spectroscopy revealed that the bottom of the parabolic dispersion was broadened but still persistent. The criterion for the occurrence of the $E(k)$ deviation was found to be given by the condition where the coherent length L_ϕ of the surface electrons becomes shorter than the wavelength of the Shockley surface-state electron.

I. INTRODUCTION

Impurity scattering plays a key role in carrier dynamics and localization in two-dimensional electron gas (2DEG) systems.[1–15] A surface state with 2DEG characteristics is the paradigm for these studies because the number of impurity atoms can be precisely controlled. The resulting change in the electronic states is directly accessible using scanning tunneling microscopy (STM), angle-resolved photoelectron spectroscopy (ARPES), and two-photon photoemission spectroscopy (2PPE).[1–8] Impurity scattering has been extensively and directly addressed using STM for single adatoms on surfaces. A resonance was reported to appear just below the Shockley surface-state band edge on Cu, Ag, Co, and Cs single adatom sites on Cu and Ag(111) surfaces.[16–19] Theoretical studies have revealed that the bound state is originated by an adatom-derived attractive potential in the two-dimensional systems.[16,17,20] However, the adatom-derived state is completely changed if the adatom is embedded in the surface layer.[21] The transition to the adatom-derived antibonding state was also reported to induce long-life unoccupied resonance for alkali-metal adatoms on Cu and Ag(111) surfaces, as determined by 2PPE and scanning tunneling spectroscopy (STS) studies.[19,22–25] Lossy scattering at the boundary was evidenced to be the dominant life-limiting process for electrons confined by adatom corrals at Ag(111) surfaces.[26,27]

At Cu(111) and (100) surfaces with a certain amount of K, Na, and Co adatoms, the lifetime of a hole in the Shockley surface state has also been reported to be substantially decreased.[9–11] The adsorbate-induced scattering and resulting decrease in lifetime have been theoretically investigated for surface-state quasiparticles.[14,15] However, the surface impurity coverage was very small and the Shockley surface state was maintained intact in these studies. At larger impurity coverage, the holes and electrons lose their coherence and their lifetime is reduced considerably, which results in the decay of the intrinsic surface band dispersion. In this case, it is of significant interest to correlate the reduction of lifetime to the decay of the surface-state dispersion with increasing impurity atoms at the surface. The (111) surface of a Cu-Al alloy has been previously studied using ARPES.[28–30] Here, the random distribution of a large amount of Al atoms is expected to decay the intrinsic Shockley surface state of the Cu(111) surface. However, the surfaces revealed long-range ordered reconstructions that had similar band dispersions to the intrinsic state. The adatom-induced surface structure was recently reported to enhance elastic scattering by the backfolding of the surface band at $p(2 \times 2)$ Cs and Na ordered overlayers on Cu(111) surfaces.[31] However, the adatoms were arranged with long-range ordering, as in the case of the Cu-Al alloy surface. The change of the Shockley surface state has also been investigated for randomly adsorbed Ag atoms on a Au(111) surface for a wide range of Ag coverage.[32–34] However, the Ag atoms merely caused a shift of the parabolic surface-state dispersion due to their electronic similarity with Au. Thus, the effect of randomly arranged impurity atoms on the surface-state dispersion has yet to be clarified.

We report a STM study on the decay of the downward parabolic dispersion by the adsorption of Bi atoms at Ag(111) surfaces for Bi coverage θ_{Bi} from 0 to 1/3 ML. Bi atoms are randomly adsorbed and take substitutional sites at the top layer of the Ag(111) surface for $\theta_{Bi} \leqslant \sim 0.3$ ML.[35] Furthermore, Bi is electronically unlike Ag. Thus, the decay of the Ag(111) Shockley surface state is expected to be observed by the random adsorption of Bi atoms in this system. The $E(k)$ relation deduced from STM dI/dV images gradually deviated from the intrinsic downward parabolic dispersion of the Ag(111) Shockley surface state at the low-energy side with increasing θ_{Bi}. However, the bottom of the parabolic dispersion was still preserved in the STS spectrum, although the onset became broader. The deviation of $E(k)$ was also observed during completion of the Bi/Ag(111) $\sqrt{3} \times \sqrt{3}$ reconstruction with the upward parabolic dispersion at $\theta_{Bi} \sim 1/3$ ML. The deviated $E(k)$ relation approached the upward parabolic dispersion of the Bi $\sqrt{3}$ surface at the high-energy side with increasing θ_{Bi}. These deviations in $E(k)$ are observed in the region where the coherent length L_ϕ of the surface electrons becomes shorter than the wavelength of the Shockley surface-state dispersion.

II. EXPERIMENT

Experiments were conducted in an ultra-high-vacuum (UHV) apparatus.[35] Ag(111) surfaces were prepared by

depositing 20-ML-thick Ag epitaxial films on Si(111) substrates.[36] Bi atoms were deposited onto the Ag(111) surface at 460 K at a rate of 0.0015 ML/s. The Bi deposition rate was evaluated by observing the extinction of the 7×7 superspots in a reflection high-energy electron diffraction (RHEED) pattern during Bi deposition onto the Si(111) 7×7 surface. The accuracy of the estimated rate was confirmed by observing the completion of the perfect Bi-induced $\sqrt{3} \times \sqrt{3}$ reconstruction on the Ag(111) surface at a Bi coverage of 1/3 ML. To estimate the Bi coverage more accurately, the Bi coverage θ_{Bi} was determined by counting the number of Bi atoms in STM images of various locations for $\theta_{Bi} \leqslant 0.1$ ML. The Bi-induced change in the surface structure and electronic state was followed by further deposition of a small amount of Bi atoms and subsequent STM observation at $0 \leqslant \theta_{Bi} \leqslant 0.333$ ML. A bias voltage V_s was applied to the sample at the temperature of supercooled liquid N_2 (65 K) during STM observations. dI/dV images and STS spectra were acquired using the conventional lock-in technique.[37] dI/dV images were obtained in the constant-height mode. The modulation voltage V_m for STS was 5 meV.

III. RESULTS AND DISCUSSION

Figure 1 shows STM and dI/dV images of the Ag(111) surfaces with adsorbed Bi. The standing-wave patterns for the pristine Ag(111) surface ($\theta_{Bi} = 0$ ML) are evolved from the step edge and misfit dislocations in the dI/dV images [Figs. 1(b) and 1(c)]. The wavelength becomes shorter with increasing electron energy E, as expected for the downward parabolic dispersion of the Ag(111) Shockley surface state.
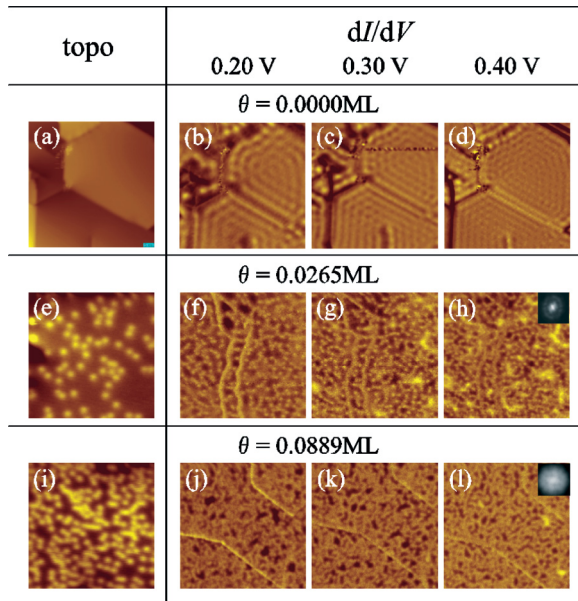
Bi atoms are located at substitutional sites on the Ag(111) surface, but are displaced upward by $\sim$0.02 nm due to their atomic size, which is larger than that of the substrate Ag atoms. Thus, the Bi atoms are observed as protrusions in the STM images.[35] During the initial stage of adsorption, the Bi atoms are adsorbed randomly, as shown in Figs. 1(e) and 1(i). At $\theta_{Bi} \leqslant \sim0.005$ ML, circular standing-wave patterns are evolved from randomly adsorbed Bi atoms, in addition to standing waves due to the step edges and misfit dislocations (not shown). At larger θ_{Bi}, scattering by Bi atoms becomes frequent, and the standing-wave patterns become more intricate, as shown in Figs. 1(f)–1(h). However, the pattern still becomes finer, i.e., the wavelength becomes shorter, with increasing E, as shown in the dI/dV images at $\theta_{Bi} = 0.0265$ ML. Finally, the wavelength became insensitive to the change in E with further increase in θ_{Bi} over ~0.08 ML, as demonstrated in Figs. 1(j)–1(l).

The wavelength λ of the surface-state electron was deduced by fast Fourier transform (FFT) of the dI/dV images. λ was then transformed to the wave vector $k = 2\pi/\lambda$. Figure 2 shows the $E(k)$ relation obtained from the dI/dV images. At $\theta_{Bi} = 0$ ML, the downward parabolic dispersion of the intrinsic Shockley surface state is reproduced, as shown by the black solid line in Fig. 2. The estimated bottom from the parabolic dispersion is located at $E = \sim0$ eV, which is consistent with previous studies that reported an upward shift of the intrinsic Ag(111) surface states at -65 meV[38] at the surface of ultrathin epitaxial Ag films grown on Si(111) substrates due to the misfit strain.[39,40] The surface state is depopulated[39] and is located at $E \sim 0$ eV for 20-ML-thick Ag films.[40] $E(k)$ deviated from the parabolic dispersion at the low-energy side with increasing θ_{Bi}, as evidenced by the blue ($\theta_{Bi} = 0.0215$ ML) and red ($\theta_{Bi} = 0.0406$ ML) solid lines in Fig. 2. The Bi-induced change

FIG. 1. (Color online) STM images (left) and dI/dV images (right) of the Ag(111) surfaces with adsorbed Bi. The STM image size is 15×15 nm^2 and that for the dI/dV images is 30×30 nm^2. The STM images were obtained at $V_s = -0.4$ V and $I_t = 0.3$ nA. The I_t used for the dI/dV images was 0.5 nA. Examples of fast Fourier transform (FFT) patterns of the dI/dV images are given as insets in (h) and (l).
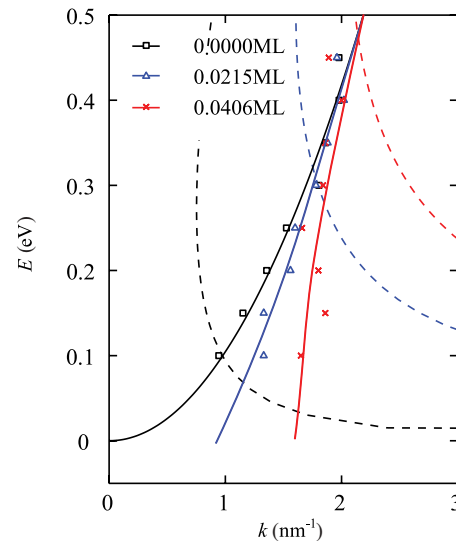

FIG. 2. (Color online) $E(k)$ relation for Ag(111) surfaces with adsorbed Bi atoms deduced by FFT of dI/dV images with $\theta_{Bi} = 0$, 0.0215, and 0.0406 ML. The black solid line is the parabolic fit of the data for $\theta_{Bi} = 0$ ML. The blue and red solid lines indicate the trend of the data plots. The dotted lines are the numerically estimated boundaries, below which the wavy nature of the surface electron is not well defined.
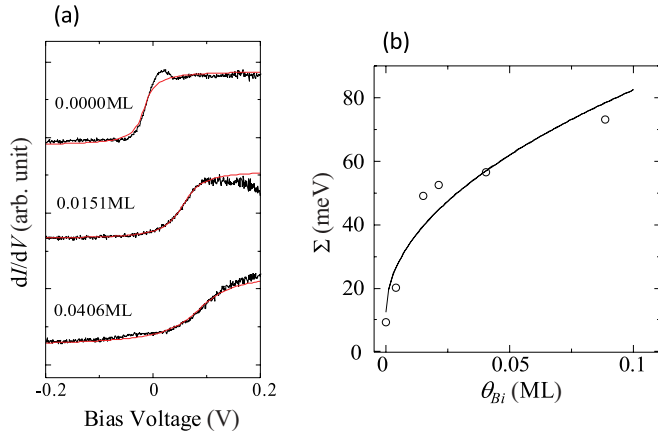
FIG. 3. (Color online) (a) STS spectra of Ag(111) surfaces with adsorbed Bi atoms for θ_{Bi} is 0, 0.015, and 0.0406 ML. (b) Self-energy Σ vs θ_{Bi}, where Σ was estimated from the width of the surface-state onset (Δ) in the STS spectrum using the equation given by Li *et al.*[1]

in $E(k)$ may appear to be the result of the downward shift of the parabolic surface dispersion. However, STS spectra revealed that the bottom of the surface state is still located above the Fermi level. Thus, $E(k)$, which was deduced from the dI/dV images, is regarded to be actually deviated from the parabolic dispersion at the low-energy side. The deviation became significant with θ_{Bi}. Finally, k became completely insensitive to the E deviation at $\theta_{Bi} \geqslant 0.08$ ML.

Figure 3(a) shows the change of the STS spectra with θ_{Bi}. The pristine ultrathin Ag(111) film surface reveals a sharp onset at the bottom of the Shockley surface state around the Fermi level, as expected for the dispersion (black solid line in Fig. 2) from the dI/dV images. However, the onset becomes broader and shifts toward the higher-energy side with increasing θ_{Bi}. Finally, the onset disappears at $\theta_{Bi} \sim 0.1$ ML (not shown). The broadening of the onset (Δ) is a measure of the reduction in the lifetime (τ) of the surface-state electrons.[1] Bi atoms scatter the surface-state electrons randomly, and the electronic state $|k\rangle$ is merged with the $|k'\rangle$ state upon scattering. This gives $|k\rangle$ an ill-defined quantum number and reduces its lifetime, τ. Thus, it is reasonable that Δ increases with θ_{Bi}. However, the persistence of the onset is inconsistent with $E(k)$ for the Bi-covered surfaces (blue and red solid lines in Fig. 2). The steplike onset originates from the two-dimensional free-electron-like dispersion at the surface. Thus, the results in Fig. 3 strongly suggest that the Bi-covered surfaces intrinsically preserve the parabolic dispersion of the Ag(111) surface for $\theta_{Bi} \leqslant \sim 0.1$ ML. In this respect, we regard the Bi-covered surface as having a parabolic dispersion, although $E(k)$, which was deduced from dI/dV images, showed an apparent deviation at the low-energy side for some reason.

A possible cause for the $E(k)$ deviation is the disturbance of the standing-wave formation by the decrease in the nearest-neighbor Bi-Bi atom distance, d_{Bi-Bi}. λ increases in the downward parabolic dispersion and could become longer than d_{Bi-Bi} at low energy. In this case, the standing-wave formation between neighboring Bi atoms would be disturbed. Thus, the dI/dV images are expected to reveal the deviation in

$E(k)$, although the surface state still preserves the downward parabolic dispersion. However, d_{Bi-Bi} is ~ 1 nm, even at small θ_{Bi}, such as 0.02 ML. Therefore, $E(k)$ should deviate for $k \leqslant 2\pi/d_{Bi-Bi} \simeq 6$ nm^{-1}, and thus, the deviation should be observed in the entire region shown in Fig. 2. However, this does not agree with the experimental results, in which $E(k)$ is only partially deviated in the low-energy region at $\theta_{Bi} = 0.0215$ and 0.0406 ML. Therefore, we suggest that d_{Bi-Bi} is not the direct criterion that determines the deviation in $E(k)$.

Alternatively, the deviation of $E(k)$ could be caused by the reduced coherent length, L_ϕ. L_ϕ is given by the product of the group velocity v_g, and the lifetime τ, of surface electrons. The STS spectra suggest that the parabolic dispersion, and thus v_g, do not exhibit a sensitive change to θ_{Bi}. However, the electron scattering at Bi atoms decreases τ. Therefore, L_ϕ is reduced with θ_{Bi}, as well as d_{Bi-Bi}. However, there is plenty of free space between Bi atoms, so that L_ϕ is reasonably expected to be longer than d_{Bi-Bi}. Thus, the criterion for the occurrence of the deviation in $E(k)$ could possibly be explained in terms of $L_\phi \leqslant \lambda$. For $L_\phi \leqslant \lambda$, the dI/dV pattern will lose a clear signature of the standing waves and the resulting $E(k)$ relation will show a deviation from the intrinsic surface dispersion. In the following, we discuss whether the criterion for the occurrence of the deviation in $E(k)$ is given by the condition $L_\phi \leqslant \lambda$. However, it is beyond the scope of this paper to theoretically reproduce the experimentally observed $E(k)$ curves in the region of $L_\phi \leqslant \lambda$, in which the dI/dV pattern is constructed by the complex scattering and mixing of surface-state electrons. It should be noted that a trend of the k independence of E was also observed for 15% Co on an InAs(110) surface.[41] At the surface with strong disorder induced by impurities, the dominating λ of the intrinsic surface dispersion[42] was suppressed in the dI/dV patterns. The FFT patterns showed filled circles due to the impurity scattering-induced mixing of the surface waves, and the radius was barely dependent on the energy, as observed in the Bi/Ag(111) system at $\theta \geqslant 0.08$ ML.

L_ϕ was evaluated to elucidate a quantitative explanation of the criterion for the occurrence of the $E(k)$ deviation. As a first step, the self-energy Σ was estimated from the STS spectra to obtain $\tau = \hbar/2\Sigma$ for the surface electrons.[43] However, v_g was difficult to estimate experimentally because $E(k)$ from the dI/dV images was regarded to deviate from the true dispersion. However, the STS spectra suggested that the Bi-covered surfaces almost preserved the intrinsic parabolic dispersion of the Ag(111) surfaces for small θ_{Bi}. Thus, in the following, v_g was roughly estimated under the assumption that the surface electrons have a downward parabolic dispersion with the same effective mass as the Ag(111) Shockley surface state, even for the Bi-adsorbed surface. $k_{crit} = 2\pi/L_\phi$ was evaluated using the estimated τ and v_g values to define the region where $E(k)$ deviates from the intrinsic Shockley surface-state dispersion.

Σ is related to the broadening Δ of the onset in the STS spectra by $\Delta \simeq \frac{\pi V_m^2}{2} \frac{1}{\sqrt{\Sigma^2 + V_m^2} - \Sigma} + O(\frac{T}{\Sigma})^2$ at the band bottom.[1] The STS spectra were fitted using an arctangent-type step function to deduce Δ, and Σ was estimated using the above equation.[44] The obtained Σ is plotted as a function of θ_{Bi} in

Fig. 3(b). Σ increases from 8 to 70 meV with increasing θ_{Bi} from 0 to 0.1 ML. The increase is approximately proportional to $\sqrt{\theta_{\mathrm{Bi}}}$.[45] Σ typically consists of three components: electron-electron scattering (Σ_{e-e}), electron-phonon scattering ($\Sigma_{e-\mathrm{ph}}$), and impurity/defect scattering (Σ_{imp}). Among the three components, Σ_{e-e} has previously been reported to govern Σ for defect-free Ag(111) wide terraces at 5 K.[1] However, $\Sigma_{e-\mathrm{ph}}$ is also expected to be activated at the present measurement temperature ($T = 65$ K). In addition, the Ag(111) surfaces of ultrathin films include misfit dislocations,[36] and Bi atoms are adsorbed randomly on the surfaces. These factors increase Σ_{imp} considerably; therefore, all three components contribute to Σ in the present system.

For defect-free Ag(111) terraces at 5 K, where Σ is dominated by Σ_{e-e}, τ was reported to reveal an almost Fermi-liquid-like dependence on E [i.e., $(E - E_F)^{-2}$].[3,4] Therefore, we adopted the *correct* coefficient 5.2 fs eV^2 for $\tau = \hbar/2\Sigma_{e-e}$ (Refs. 46 and 47) to evaluate $\Sigma_{e-e}(E)$ as a function of E. The resulting Σ_{e-e} is increased from 0 to 16 meV with increasing E from 0 to 0.5 eV. It should be noted that we are not aware of any previous studies on the E dependence for $\Sigma_{e-\mathrm{ph}}$ and Σ_{imp} at Ag(111) surfaces; therefore, the E dependence is neglected as a crude approximation. However, for the electron-phonon contribution, $\Gamma = 2\Sigma_{e-\mathrm{ph}}$ was reported to increase linearly with T at the Cu(111) surface.[6] The electron-phonon coupling constant was estimated to be $\Lambda = 0.14$ at the Cu(111) surface from the linear coefficient (0.0704 meV/K) of Γ ($= 2\Lambda kT$). $\Lambda = 0.13$ was adopted for the Ag(111) surface,[1] and $\Sigma_{e-\mathrm{ph}}$ was estimated to be ~ 2 meV at $T = 65$ K. Finally, Σ_{e-e} and $\Sigma_{e-\mathrm{ph}}$ were subtracted from Σ at the band bottom [Fig. 3(b)], and assigned the difference as Σ_{imp}. $\Sigma_{e-\mathrm{ph}}$ and Σ_{e-e} are less than 20 meV, which indicates that Σ is dominated by Σ_{imp}.

Σ, L_ϕ, and $k_{\mathrm{crit}} = 2\pi/L_\phi$ were calculated as functions of E and θ_{Bi} using these $\Sigma_{e-e}(E)$, $\Sigma_{e-\mathrm{ph}}$, and Σ_{imp}. The resulting $k_{\mathrm{crit}}(E)$ is indicated by dashed lines for $\theta_{\mathrm{Bi}} = 0$, 0.0215, and 0.0406 ML in Fig. 2. Under the dashed line, k becomes smaller than k_{crit}. Thus, $E(k)$ from the dI/dV images are expected to indicate the deviation in the region below the intersection of k_{crit} (dashed line) and the intrinsic parabolic surface dispersion (black solid line). The deviation of $E(k)$ did occur in this region for both $\theta_{\mathrm{Bi}} = 0.0215$ and 0.0406 ML. Therefore, we consider the criterion for the deviation of $E(k)$ is quantitatively given by the relation $L_\phi \leqslant \lambda$ for the Ag(111) surface with randomly adsorbed Bi atoms.

The entire downward parabolic dispersion enters the complete region below the k_{crit} line at $\theta_{\mathrm{Bi}} \sim 0.08$ ML (not shown). We consider that this is the reason why $E(k)$ became insensitive to E at this coverage. However, the surface state was still persistent, although the onset was significantly broadened in the STS spectrum at this coverage (not shown). The Shockley surface state is regarded to be completely collapsed at $\theta_{\mathrm{Bi}} \geqslant 0.1$ ML from the disappearance of the onset in the STS spectrum.

The deviation in $E(k)$ was also observed during the course of completion of the Bi-induced $\sqrt{3} \times \sqrt{3}$ reconstruction at the Ag(111) surface. At $\theta_{\mathrm{Bi}} \sim 1/3$ ML, randomly distributed Bi atoms possess $\sqrt{3} \times \sqrt{3}$ long-range ordering. The characteristic upward parabolic dispersion of the Bi/Ag(111) $\sqrt{3} \times \sqrt{3}$ surface[37] was gradually completed during this stage.
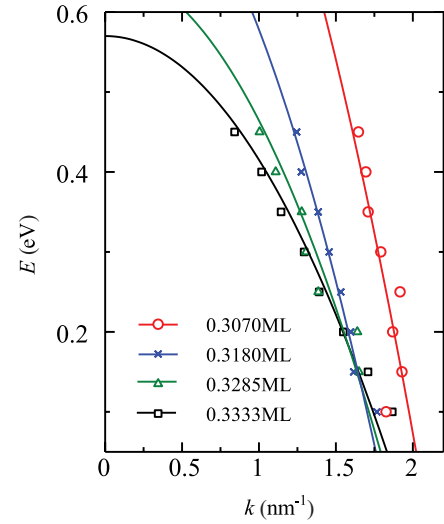

FIG. 4. (Color online) $E(k)$ relation of the Ag(111) surface with adsorbed Bi atoms at θ_{Bi} close to 0.33 ML. θ_{Bi} is indicated in the key legend. k was deduced by FFT analysis of the dI/dV images. The black curve is the parabolic fitting of the data at $\theta_{\mathrm{Bi}} = 0.3333$ ML. Other fitting curves indicate the trend of the data plots.

Figure 4 represents the θ_{Bi}-dependent change of $E(k)$ at $\theta_{\mathrm{Bi}} \sim 1/3$ ML. Contrary to the decay of the Ag(111) Shockley surface state in the initial stage, the $E(k)$ relation showed a deviation at the *high*-energy side. The deviation became smaller and $E(k)$ approached the intrinsic upward parabolic dispersion with θ_{Bi}. For the Bi-induced $\sqrt{3} \times \sqrt{3}$ surface band electrons, unsubstituted Ag atoms serve as scattering atoms at $\theta_{\mathrm{Bi}} \sim 1/3$ ML. Thus, the increase in θ_{Bi} causes a decrease in the number of scattering sites for the upward parabolic surface-state electrons at the Bi/Ag(111) $\sqrt{3} \times \sqrt{3}$ surface. The high-energy side is close to the edge of the upward parabolic band dispersion; therefore, by the same reason for the deviation of $E(k)$ from the Ag(111) Shockley surface-state dispersion at low θ_{Bi}, the $k \leqslant k_{\mathrm{crit}}$ region is expected to appear at the high-energy side with increasing λ and decreasing L_ϕ. This $k \leqslant k_{\mathrm{crit}}$ region shrinks with θ_{Bi} because the decrease in the remaining Ag atom impurities increases the lifetime τ. As a result, the deviation is reduced and $E(k)$ approaches the intrinsic upward parabolic dispersion of the Bi/Ag(111) $\sqrt{3} \times \sqrt{3}$ surface with θ_{Bi}.

IV. SUMMARY

In summary, the decay of the downward parabolic Shockley surface-state dispersion by the random adsorption of Bi atoms on the Ag(111) surface was investigated. The $E(k)$ relation gradually deviated from the intrinsic dispersion at the low-energy side with θ_{Bi}. However, the onset of the Shockley surface state became broader, although persistent, for $\theta_{\mathrm{Bi}} \leqslant \sim 0.1$ ML. The deviation in $E(k)$ was attributed to a reduction of L_ϕ with θ_{Bi}. The quantitative criterion was given by the condition $L_\phi \leqslant \lambda$. This condition is satisfied, specifically at the low-energy side, due to the decrease in v_g and increase in λ near the band edge. The deviation was extended due to the decrease in τ with θ_{Bi}. The deviation of $E(k)$ was also observed in the course of the completion of the Bi/Ag(111) $\sqrt{3} \times \sqrt{3}$

reconstruction at $\theta_{Bi} \sim 1/3$ ML. The deviation appeared at the high-energy side and was reduced with increasing θ_{Bi}. This deviation of $E(k)$ was also regarded to occur due to $L_\phi \leqslant \lambda$ near the band edge.

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[1] J. Li, W-D. Schneider, R. Berndt, O. R. Bryant, and S. Crampin, Phys. Rev. Lett. **81**, 4464 (1998).

[2] J. Kliewer, R. Brendt, E. V. Chulkov, V. M. Silkin, P. M. Echenique, and S. Crampin, Science **288**, 1399 (2000).

[3] L. Bürgi, O. Jeandupeux, H. Brune, and K. Kern, Phys. Rev. Lett. **82**, 4516 (1999).

[4] L. Vitali, P. Wahl, M. A. Schenider, K. Kern, V. M. Silkin, E. V. Chulkov, and P. M. Echenique, Surf. Sci. Lett. **523**, L47 (2003).

[5] S. D. Kevan, Phys. Rev. Lett. **50**, 526 (1983).

[6] B. A. McDougall, T. Balasubramanian, and E. Jensen, Phys. Rev. B **51**, 13891 (1995).

[7] W. Berthold, U. Höfer, P. Feulner, E. V. Chulkov, V. M. Silkin, and P. M. Echenique, Phys. Rev. Lett. **88**, 056805 (2002).

[8] K. Boger, M. Roth, M. Weinelt, Th. Fauster, and P.-G. Reinhard, Phys. Rev. B **65**, 075104 (2002).

[9] S. D. Kevan, Phys. Rev. B **33**, 4364 (1986).

[10] X. Y. Wang, R. Paiella, and R. M. Osgood, Jr., Phys. Rev. B **51**, 17035 (1995).

[11] M. Hirschiman and T. Fauster, Appl. Phys. A **88**, 547 (2007).

[12] J. Wiebe, Chr. Meyer, J. Klijn, M. Morgenstern, and R. Wiesendanger, Phys. Rev. B **68**, 041402 (2003).

[13] M. Morgenstern, J. Klijn, Chr. Meyer, M. Getzlaff, R. Adelung, R. A. Römer, K. Rossnagel, L. Kipp, M. Skibowski, and R. Wiesendanger, Phys. Rev. Lett. **89**, 136806 (2002).

[14] S. Crampin, M. H. Boon, and J. E. Inglesfield, Phys. Rev. Lett. **73**, 1015 (1994).

[15] B. Gumhalter, A. Šiber, H. Buljan, and T. Fauster, Phys. Rev. B **78**, 155410 (2008).

[16] F. E. Olsson, M. Persson, A. G. Borisov, J.-P. Gauyacq, J. Lagoute, and S. Fölsch, Phys. Rev. Lett. **93**, 206803 (2004).

[17] L. Limot, E. Pehlke, J. Kröger, and R. Berndt, Phys. Rev. Lett. **94**, 036805 (2005).

[18] N. Néel, J. Kröger, R. Berndt, and E. Pehlke, Phys. Rev. B **78**, 233402 (2008).

[19] M. Ziegler, J. Kröger, R. Berndt, A. G. Borisov, and J. Gauyacq, Phys. Rev. B **79**, 075401 (2009).

[20] E. Zaremba, I. Nagy, and P. M. Echenique, Phys. Rev. Lett. **90**, 046801 (2003).

[21] S. Lounis, P. Mavropoulos, P. H. Dederichs, and S. Blügel, Phys. Rev. B **73**, 195421 (2006).

[22] N. Fischer, S. Schuppler, Th. Fauster, and W. Steinmann, Surf. Sci. **314**, 891 (1994).

[23] M. Bauer, S. Pawlik, and M. Aeschlimann, Phys. Rev. B **55**, 10040 (1997).

[24] S. Ogawa, H. Nagano, and H. Petek, Phys. Rev. Lett. **82**, 1931 (1999).

[25] M. Bauer, S. Pawlik, and M. Aeschlimann, Phys. Rev. B **60**, 5016 (1999).

[26] H. Jensen, J. Kröger, R. Berndt, and S. Crampin, Phys. Rev. B **71**, 155417 (2005).

[27] S. Crampin, H. Jensen, J. Kröger, L. Limot, and R. Berndt, Phys. Rev. B **72**, 035443 (2005).

[28] H. Asonen, M. Lindroos, M. Pessa, R. Prasad, R. S. Rao, and A. Bansil, Phys. Rev. B **25**, 7075 (1982).

[29] R. Courths, M. Lau, T. Scheunemann, H. Gollisch, and R. Feder, Phys. Rev. B **63**, 195110 (2001).

[30] Y. Yu, K. Sagisaka, and D. Fujita, Phys. Rev. B **79**, 235427 (2009).

[31] C. Corriol, V. M. Silkin, D. Sánchez-Portal, A. Arnau, E. V. Chulkov, P. M. Echenique, T. von Hofe, J. Kliewer, J. Kröger, and R. Berndt, Phys. Rev. Lett. **95**, 176802 (2005).

[32] H. Cercellier, Y. Fagot-Revurat, B. Kierren, F. Reinert, D. Popovič, and D. Malterre, Phys. Rev. B **70**, 193412 (2004).

[33] F. J. Palomares, M. Serrano, A. Ruiz, F. Soria, K. Horn, and M. Alonso, Surf. Sci. **513**, 283 (2002).

[34] A. Bendounan, H. Cercellier, Y. Fagot-Revurat, B. Kierren, V. Y. Yurov, and D. Malterre, Phys. Rev. B **67**, 165412 (2003).

[35] C. Kato, Y. Aoki, and H. Hirayama, Phys. Rev. B **82**, 165407 (2010).

[36] M. Miyzaki and H. Hirayama, Surf. Sci. **602**, 276 (2008).

[37] H. Hirayama, Y. Aoki, and C. Kato, Phys. Rev. Lett. **107**, 027204 (2011).

[38] M. Becker, S. Crampin, and R. Berndt, Phys. Rev. B **73**, 081402(R) (2006).

[39] G. Neuhold and K. Horn, Phys. Rev. Lett. **78**, 1327 (1997).

[40] K. Sawa, Y. Aoki, and H. Hirayama, Phys. Rev. B **80**, 035428 (2009).

[41] J. Wiebe, Chr. Meyer, J. Klijn, M. Morgenstern, and R. Wiesendanger, Phys. Rev. B **68**, 041402 (2003).

[42] M. Morgenstern, J. Klijn, Chr. Meyer, M. Getzlaff, R. Adelung, R. A. Römer, K. Rossnagel, L. Kipp, M. Skibowski, and R. Wiesendanger, Phys. Rev. Lett. **89**, 136806 (2002).

[43] Correctly, $\tau = \beta \times \hbar/2\Sigma$, where β is a factor that represents the extent to which the tunneling state couples to lifetime-limiting processes. For Ag(111) surfaces, β has been reported to be close to one (see Ref. 1). Therefore, in the discussion, we have assumed that $\beta = 1$ for the Bi-covered Ag(111) surfaces, although the effect of a small amount of Bi on β has not been reported.

[44] For the obtained Σ, $(T/\Sigma)^2$ is moderately small so as to be regarded negligible at the measurement temperature of $T = 65$ K ($\sim$5 meV); therefore, the estimation is considered to be meaningful.

[45] H-J. Noh, J. Jeong, E-J. Cho, H-K. Lee, and H-Y. Kim, Europhys. Lett. **96**, 47002 (2011).

[46] S. Crampin, J. Kröger, H. Jensen, and R. Berndt, Phys. Rev. Lett. **95**, 029701 (2005).

[47] J. Kröger, L. Limot, H. Jensen, R. Brendt, E. Pehlke, and S. Crampin, Prog. Surf. Sci. **80**, 26 (2005).