Experimental observation of vibrational modes on Ag(111) along ΓM and ΓK

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We present an off-specular high-resolution electron energy-loss spectroscopy study of the vibrational modes along the $\overline{\Gamma M}$ and $\overline{\Gamma K}$ directions in the surface Brillouin zone of clean Ag(111). We show experimental data of vibrational modes at the $\overline{K}$ edge, thereby extending the earlier He-scattering data on the S_1 Rayleigh mode and pseudo-Rayleigh mode. In addition, we observed the S_3 gap mode and three high-frequency modes along $\overline{\Gamma K}$. According to our knowledge, this is the first experimental evidence for the existence of these high-frequency modes in the (111) surface Brillouin zone of fcc metals. Along $\overline{\Gamma M}$ the experimental dataset of the S_2 gap mode is extended to smaller $Q_{//}$, and for the first time the high-frequency vibrational mode close to the top of the projected bulk phonon band is observed. The EELS data is analyzed by comparison to theoretical studies previously reported in the literature. This paper presents new experimental data of the surface vibrational structure,which can stimulate theoreticians to refine their modeling and improve the understanding of the Ag(111) surface lattice dynamics.

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I. INTRODUCTION

One of the central issues in surface science is to understand how the static and dynamic properties of a surface differ from those in the bulk. The dynamical properties of a surface can be explored experimentally by probing the surface vibrational modes, which are directly related to the interatomic force constants. Despite the considerable interest in the surface lattice dynamics of Ag(111), the number of experimental studies that report about its surface vibrational structure is limited. Experimental information is essential to verify surface lattice dynamical descriptions and to improve the detailed understanding of interaction potentials at surfaces, as illustrated below by the short description of the historical development of the current understanding of the Ag(111) surface lattice dynamics.

In the first half of the eighties helium-atom scattering (HAS) studies of Ag(111) were reported by Yerkes and Miller[1] and Doak *et al.*[2] The latter study reported the experimental observation of the Rayleigh mode and a resonance mode along both the $\overline{\Gamma M}$ and $\overline{\Gamma K}$ directions,which at that time could only be explained by assuming anomalous softening (48%) of the intralayer radial force constant using a force constant model.[3] Various approaches[4–8] were reported to explain the large softening, which is not expected on the unreconstructed and unrelaxed close-packed Ag(111) surface. In 1993, Chen *et al.*[9] reported a parameter-free first-principles phonon calculation and theoretically reproduced the existing HAS and electron energy-loss spectroscopy (EELS) data on Cu(111) and Ag(111) without the need to evoke exotic models, such as anomalous surface dynamical effects, on these densely packed surfaces. These studies demonstrated the need for highly accurate theoretical models for proper characterization of surface lattice dynamics on even simple metallic surfaces. In 1996, the gap mode S_2 was observed with EELS at the $\overline{M}$ edge in the surface Brillouin zone of Ag(111),[10] as predicted earlier by Chen *et al.*[9] In this EELS study only the vibrational modes along the $\overline{\Gamma M}$ direction were examined. As no other experimental studies have been reported, a large part of the vibrational modes in the surface Brillouin zone of clean Ag(111) is still unexplored, in particular along the $\overline{\Gamma K}$ direction. Along this direction the situation is more complicated than along $\overline{\Gamma M}$ due to the lack of a mirror plane, which results in a mixture of shear-horizontal and sagittal plane modes[9] and in addition the so-called pseudo-surface mode or generalized Rayleigh mode is present.[8,11]

In this paper we present an off-specular high-resolution EELS study at room temperature of the vibrational modes along the $\overline{\Gamma M}$ and $\overline{\Gamma K}$ directions in the surface Brillouin zone of clean Ag(111). Due to our relatively high instrumental resolution and good signal-to-noise ratio, we were able to separate closely-spaced phonon contributions and expand the existing set of experimental data in literature, in particular in the direction $\overline{\Gamma K}$. In both directions, the EELS data is in good agreement with the earlier reported HAS and EELS data. The experimental data is analyzed by comparison to the results of theoretical models reported in the literature, and we show that at the high-symmetry points $\overline{M}$ and $\overline{K}$ the obtained EELS data is in good agreement with these studies. In particular, the EELS data is compared to the DFT-LDA calculations of the vibrational structure of the surface Brillouin zone by Xie *et al.*[12] Note that models that predict anomalous softening of the intralayer force constants are not considered in this paper, for these were shown to be inadequate.[9,10]

II. EXPERIMENTAL

The surface of the Ag(111) crystal (MaTeck, orientation accuracy $\leq 0.5°$) was prepared by standard sputtering and anneal cycles ($T = 750$ K). The cleanliness and structure of the surface were checked with HREELS and low energy electron diffraction (LEED), respectively. The HREELS

scattering plane, the so-called sagittal plane defined by the Ag(111) surface normal and the direction of the incident wave vector, was directed along the $\overline{\Gamma M}$ and $\overline{\Gamma K}$ directions within 2° using *in situ* azimuthal sample rotation and LEED. HREELS measurements were performed in a newly constructed ultrahigh vacuum setup ($p_{\text{base}}=3\times10^{-10}$) mbar using a spectrometer (ELS3000, LK Technologies) with a double-pass monochromator and analyzer. Phonon dispersion curves were determined at room temperature along the symmetry directions of the Ag(111) surface Brillouin zone by recording off-specular HREELS spectra as a function of the incident angle of the electron beam using various electron impact energies (50, 75, 100, 125 eV). HREELS spectra were recorded by averaging typically eight repetitive scans, which were measured with energy steps of 0.178 meV/channel and an accumulation time of 1.5 s/channel. The resolution of the off-specular measurements ranged from 2.8 to 3.1 meV. In specular reflection geometry typically a resolution of 1.4 meV (FWHM of elastic-peak) was obtained. The measurement of a dispersion curve was completed within 15 h after which still no contamination was detectable with HREELS.

III. RESULTS AND DISCUSSION

Figure 1 shows the EELS spectra recorded along the $\overline{\Gamma K}$ direction as a function of the incident angle θ_i and electron impact energy. The angle of the scattered electron beam θ_s was kept constant at $\sim$81.6°. Both θ_i and θ_s are defined with respect to the surface normal. In all used scattering geometries θ_i was smaller than θ_s, which means that negative (positive) parallel momentum $Q_{//}$ was transferred from the electron to the vibrational mode upon creation (annihilation) of a phonon. The sharp features in the spectra are due to the annihilation (energy-gain) and creation (energy-loss) of surface vibrational modes caused by single-phonon inelastic processes as discussed by Roundy and Mills.[13] The peaked contributions around zero energy-loss are caused by elastically scattered electrons.

Figure 1 shows that the cross section for excitation of a certain vibrational mode depends strongly on the scattering geometry and electron impact energy. For instance, near the surface Brillouin zone $\bar{K}$ edge (e.g., lowest spectra in the graphs) the peaks denoted with * and # can only be observed using an impact energy of 50 and 75 eV, respectively. In addition, the shear-vertical (SV) polarized Rayleigh mode S_1 (denoted with arrows in Fig. 1) is detectable at all impact energies, yet its intensity typically decreases upon decreasing off-specular angle.The dependence of the EELS scattering cross section on geometry and impact energy has been reported before in experimental studies[10,14–16] and theoretical discussions of large-angle impact-scattering, which include an explicit description of the multiple scattering of both incoming and outgoing electrons from the crystal.[17,18] To detect most of the present surface vibrational modes, suitable impact energies were chosen to enhance the scattering cross sections of modes that are otherwise too low in intensity to be detected.

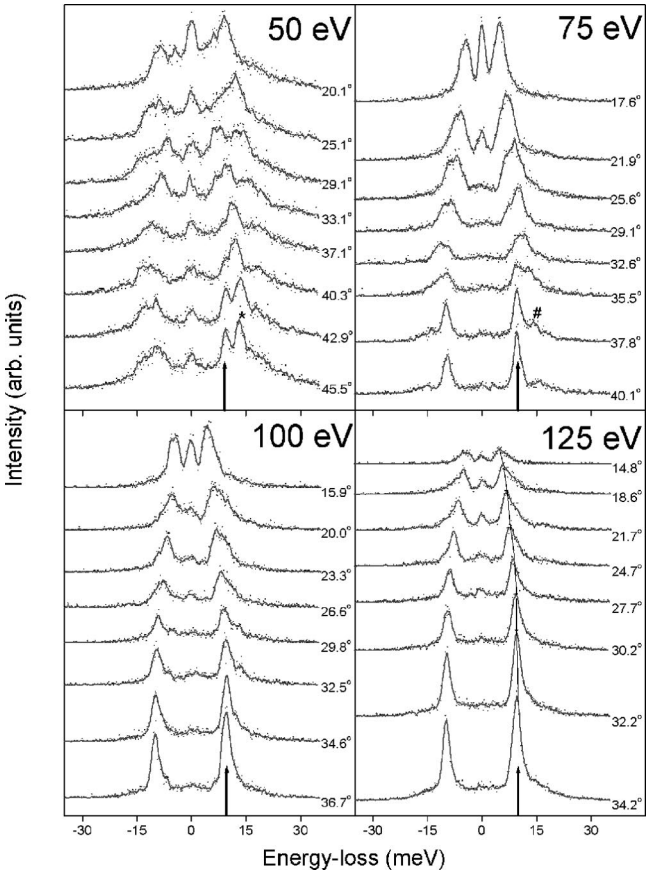

FIG. 1. EELS spectra taken along the $\overline{\Gamma K}$ direction on Ag(111) as a function of the incident angle of the incoming electron beam. The spectra are recorded with $\theta_s\sim81.6°$ and electron impact energies of 50, 75, 100, and 125 eV. For each spectrum the difference between θ_i and θ_s is denoted ($\theta_i<\theta_s$). In each graph the spectrum with the largest off-specular angle ($\theta_s-\theta_i$) represents the phonon-structure near the K edge of the Ag(111) surface.

The dependence of the peak-positions on parallel momentum transfer (e.g., incident angle) illustrates the dispersive behavior of the observed vibrational modes. Figure 2 shows the measured dispersion relations of vibrational modes along the $\overline{\Gamma M}$ and $\overline{\Gamma K}$ directions in the Ag(111) surface Brillouin zone. The data along $\overline{\Gamma M}$ was obtained by off-specular EELS measurements using impact energies of 50 and 75 eV. Due to the uncertainty in θ_s the absolute position of the dispersion curves in the surface Brillouin zone is only accurate within 0.04 to 0.02 Å^{-1} for $Q_{//}$ ranging from 1.45 to 0.40 Å^{-1}, respectively. The vibrational energies were determined by a least-squares fit of the EELS spectra with a superposition of Gaussian distributions. The FWHM of the Gaussians was fixed and set equal to that of the elastic peak. The zero of the energy-scale of a spectrum was set using the elastic peak or, when the elastic intensity was too low, the symmetry of gain- and loss-peak energies. In the case of closely spaced contributions, those contributions accurately obtainable from spectra taken with different impact energies were fixed to determine the energy of the other features more precisely. To avoid assignment of bulk bands, only those features were taken into account that, within a series re-

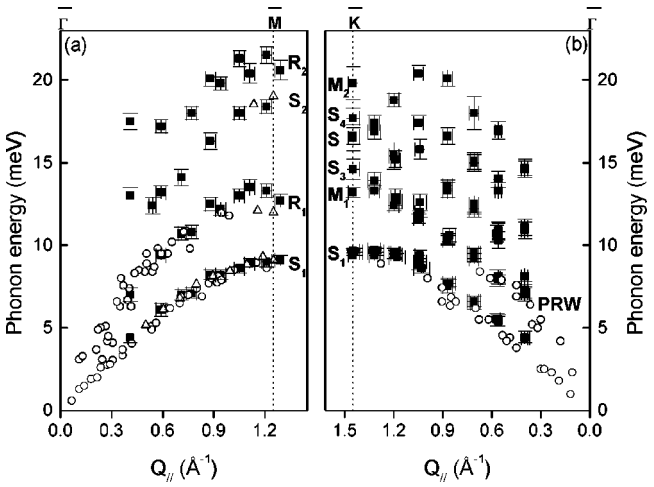

FIG. 2. Experimentally determined dispersion of vibrational modes on the Ag(111) surface along the directions (a) $\overline{\Gamma M}$ and (b) $\overline{\Gamma K}$. The filled squares denote the experimental data observed with EELS in this study. The uncertainty in $Q_{//}$ is due to the finite acceptance angle ($\sim 0.5°$) of the monochromator and analyzer of the spectrometer. The open circles [HAS (Ref. 2)] and open triangles [EELS (Ref. 10)] denote the existing set of experimental data in literature. The dotted lines in (a) and (b) give the position of the $\overline{M}$ edge and $\overline{K}$ edge, respectively.

corded with a certain impact energy, manifest themselves as clear peaks with respect to the background. As an example Fig. 3 shows the Gaussian fit of a measured spectrum. Only the four Gaussian contributions with lowest energy loss were attributed to the excitation of surface phonons. The other two

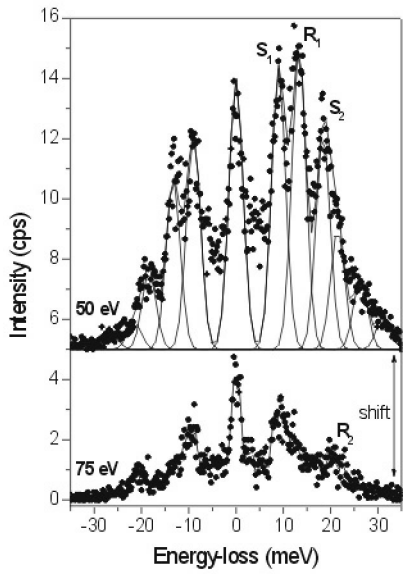

FIG. 3. Energy-loss spectra recorded near the $\overline{M}$ edge of the Ag(111) surface Brillouin zone using an impact energy of 50 and 75 eV and θ_i of, respectively, 40.6° and 44.2° ($\theta_s = 80.7°$). The spectrum taken with 50 eV is shifted vertically for clarity.The thick solid curve in the 50 eV spectrum denotes the fit consisting of multiple shown Gaussian contributions. The solid curve in the 75 eV spectrum denotes the smoothed data.

Gaussians were used to account for the background.

As illustrated in Fig. 2, the agreement between the experimental data of this study and the reported HAS (Ref. 2) and EELS (Ref. 10) data in literature is excellent in both the $\overline{\Gamma M}$ and $\overline{\Gamma K}$ direction. Concerning the $\overline{\Gamma M}$ direction and with respect to the experimental data reported in literature, the dispersion relation of the gap mode S_2 near $\overline{M}$ was extended to smaller $Q_{//}$ and the high-frequency vibrational mode R_2 close to the top of the bulk phonon band was observed. To assign this mode, the EELS data at the $\overline{M}$ edge of the surface Brillouin zone is compared to the results of theoretical studies reported in literature (see Table I). Theoretical studies that predict the anomalous softening of the intralayer surface force constants are excluded from the comparison, as this has been proven to be incorrect.[9,10] Table I shows an excellent agreement between the theoretically predicted vibrational frequencies and the experimental data. Accordingly, the high-frequency mode is attributed to be the predicted surface resonance (labeled R_2), which has a small first-layer shear-vertical (SV) polarization and a large second-layer longitudinal polarization.[9,19,20] Near and at $\overline{M}$ a small discrepancy (~ 1 meV) is noted between the measured surface resonance R_1 and the EELS data in the literature.[10] Comparison of our spectra (see Fig. 3) to the spectrum from which Chen *et al.* derived the frequency of the R_1 mode (Fig. 2 in Ref. 10) shows, that the R_1 feature observed by Chen *et al.* actually consists of multiple phonon contributions, namely, the S_1 and R_1 (and S_2) modes. As the current EELS study fully resolves these contributions, the R_1-mode frequencies near and at $\overline{M}$ reported in the present paper are considered to be more accurate.

Along $\overline{\Gamma K}$, the measured Rayleigh mode S_1 and SV-polarized pseudo-Rayleigh mode (PRW),[8,9] are in excellent agreement with the HAS data reported by Doak *et al.*[2] The present EELS study extends the earlier He-scattering work out to $\overline{K}$, thereby showing a splitting of the pseudo-Rayleigh mode into two branches near the $\overline{K}$ edge. In addition, we report the observation of three high-frequency modes on Ag(111) along $\overline{\Gamma K}$. To assign the observed vibrational modes at the $\overline{K}$ edge, the experimental data is compared to theoretically predicted frequencies (Table I), including the calculated vibrational structure along $\overline{\Gamma K}$ by Xie *et al.*[12] (see Fig. 4). Comparison of the splitted PRW near $\overline{K}$ with the results reported by Xie *et al.* suggests, that the upper-frequency branch is the primarily shear-horizontal (SH) polarized gap mode S_3 and the lower-frequency branch (labeled M_1) is located in the projected bulk-phonon density of states. Moreover,the mode observed at 4.0 ± 0.1 THz is tentatively assigned to be the predicted gap mode (labeled S),[12] which is located above S_3 and just below the bulk band. Based on the agreement between the mode observed at 4.3 ± 0.1 THz and both the FP and DFT-LDA calculations, we suggest this mode to be the quasilongitudinal (SP) polarized mode S_4. The observation of the high-frequency vibrational mode close to the top of the bulk band is consistent with the enhanced vibrational density of states predicted by Xie *et al.*

TABLE I. Comparison of the experimental and theoretical vibrational frequencies (THz) at the high-symmetry points $\overline{M}$ and $\overline{K}$ in the surface Brillouin zone of Ag(111). If known, the displacement polarization of the atoms in the topmost surface layer is given in brackets.

M edge	S_1 (SV)	R_1 (SV)	S_2 (SP)	R_2 (SV)		
this study	2.19 ± 0.02	3.1 ± 0.1	4.4 ± 0.1	5.1 ± 0.1		
FP (Ref. 9)	2.2	3.0 ± 0.1 [a]	4.5 ± 0.1 [a]	4.8 ± 0.1 [a]		
MD/EAM (Ref. 19)	2.15	3.2 ± 0.1 [b]	4.33	4.9 ± 0.1 [b]		
MD/EAM (Ref. 20)	2.2 ± 0.1 [c]	3.4 ± 0.1 [c]	4.4 ± 0.1 [c]	4.9 ± 0.1 [c]		
QHA/EAM (Ref. 20)	2.1 ± 0.1 [c]		4.3 ± 0.1 [c]			
DFT-LDA (Ref. 12)	2.16 ± 0.05 [d]		4.58 ± 0.05 [d]			
FPhonon/EAM (Ref. 21)	2.3		4.7			
K edge	S_1 (SV)	M_1	S_3 (SH)	S	S_4 (SP)	M_2
this study	2.31 ± 0.02	3.19 ± 0.07	3.5 ± 0.1	4.0 ± 0.1	4.3 ± 0.1	4.8 ± 0.1
FP (Ref. 9)	2.1				4.3 ± 0.1 [e]	
DFT-LDA (Ref. 12)	2.30 ± 0.05 [d]		3.70 ± 0.02 [d]	3.88 ± 0.02 [d]	4.42 ± 0.02 [d]	

[a]Determined from Fig. 3 in Ref. 9.
[b]Determined from Fig. 2 in Ref. 19.
[c]Determined from Fig. 3 in Ref. 20.
[d]Determined from Fig. 7 in Ref. 12.
[e]Determined from Fig. 4 in Ref. 21.

The above discussion shows a good agreement between the theoretical results and the experimental data at the high-symmetry points, in particular at $\overline{M}$, of the Ag(111) surface Brillouin zone. Although at high-symmetry points the dependence of vibrational frequencies on the surface force field is the largest,[22] a valid theoretical description of the surface lattice dynamics should also predict correctly the dispersion of vibrational modes. Figure 4 compares the EELS data to the vibrational structure of the complete surface Brillouin zone calculated by Xie *et al.*[12] using density-functional perturbation theory within the local-density approximation (DFT-LDA). In addition to these calculations, Chen *et al.*[9] reported some phonon energies midway the $\overline{\Gamma M}$ direction of the S_1 mode (1.5 THz), R_1 mode (2.5 THz), and longitudinally polarized mode (3.4 THz), which are consistent with our experimental data.

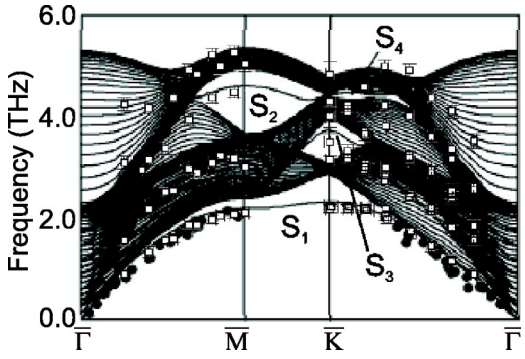

FIG. 4. Comparison between the obtained EELS data and the vibrational structure of the surface Brillouin zone calculated by Xie *et al.* (Ref. 12) using density-functional perturbation theory within the local-density approximation (DFT-LDA) (reprinted with permission).The solid circles denote the experimental data from He scattering.

According to Fig. 4, the *ab initio* DFT-LDA calculations of the surface vibrational structure and the experimental data are overall in good agreement. The most remarkable are (i) the excellent agreement between theory and experiment concerning the SV-polarized Rayleigh mode S_1 along both $\overline{\Gamma M}$ and $\overline{\Gamma K}$ and (ii) the deviation between the calculated and experimental data of the gap modes S_2 and S_3. The excellent agreement between the experimental and theoretical dispersion of the Rayleigh mode S_1 suggest, that the force constants that couple the first- and deeper-layer atoms are well described by the DFT-LDA calculations, as the S_1 mode is very sensitive to these force constants normal to the surface[22] and insensitive to the intraplanar force constants.[15] Zone-boundary gap modes, such as S_2 and S_3, are in general sensitive to the force constants, and the crucial role played by such modes in elucidating the nature of the surface lattice dynamics has been demonstrated.[23] In particular, the large sensitivity of the longitudinal (SP) polarized S_2 mode on the intraplanar force constants has been reported.[10,15,22,23] As the S_3 gap mode is primarily horizontally (SH) polarized a high sensitivity for intraplanar force constants can be expected. Therefore, we suggest that the discrepancy between the theoretical and experimental dispersion of the S_2 and S_3 gap modes points out a shortcoming in the description of the intraplanar force constants on the Ag(111) surface by the *ab initio* DFT-LDA calculations. Hence, the new EELS data provides the possibility to refine the modeling and to improve the understanding of the Ag(111) surface lattice dynamics.

In the last decade, many papers were reported[12,19–21,24,25] in an attempt to explain the (controversial) experimental observation of the anomalous thermal surface expansion of Ag(111).[26] It has been shown that a proper sampling of the vibrational modes over the surface Brillouin zone is neces-

sary to obtain a quantitatively correct description of the thermal surface expansion of Ag(111).[12,21] Xie *et al.* used their DFT-LDA results to calculate the thermal surface expansion of Ag(111) and reported, that the enhanced thermal expansion at the Ag(111) surface is governed by the anharmonicity of the interlayer potential normal to the surface as well as the softening of the parallel modes with increasing interlayer separation.[12] In particular, the importance of the softening of the parallel polarized modes relative to that of the perpendicular polarized modes was emphasized. Hence, the observed shortcomings in the description of the surface lattice dynamics may have influenced the calculated thermal surface expansion of the Ag(111) surface.

IV. CONCLUSIONS

We have presented an off-specular high-resolution electron energy-loss study of the surface vibrations along the $\overline{\Gamma M}$ and $\overline{\Gamma K}$ directions in the surface Brillouin zone of clean Ag(111) at room temperature. In both directions, the obtained EELS data is in good agreement with the existing set of experimental data in literature. We observed vibrational modes at the $\bar{K}$ edge, and along $\overline{\Gamma K}$ we have identified the S_3 gap mode and measured in addition three new high-frequency modes. To our knowledge, this is the first experimental evidence for the existence of these high-frequency modes on the (111) surface of fcc metals. Along $\overline{\Gamma M}$, we observed the high-frequency surface resonance.

At the high-symmetry points, the EELS data is in good agreement with the results of previously reported theoretical studies. In particular, we have shown that the *ab initio* DFT-LDA calculations by Xie *et al.*[12] of the Ag(111) surface vibrational structure provide a good description of the experimental data, even though there are some discrepancies concerning the dispersion of the S_2 and S_3 gap modes. Although this paper does not provide the necessary temperature-dependent data on the anharmonicity of the vibrational modes to solve the issue of the thermal surface expansion of Ag(111), we suggest that with the large amount of experimental data, in particular the observed S_3 gap mode and extended dataset on the S_2 gap mode, the description of the surface lattice dynamics may be improved and with that the description of the thermal surface expansion.

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