ordered state, in the absence of any adjustable parameters, the agreement is satisfactory. There are small quantitative differences for T < 0.15 μK. The experimental susceptibility while following the predicted saturation, appears a little higher. The spin entropy apparently settles into a $(\ln T)$-type temperature dependence. Another possible source of fluctuations is the presence of the two isotopes ^{63}Cu and ^{65}Cu (abundance ratio of 7:3) with different gyromagnetic ratios (difference of ~ 7%). In a cubic lattice, the dipolar field is zero only for a uniform environment. The spatially isotope distribution corresponds to a random magnetic field. We plan to study this effect later in some detail.[13]

In conclusion, metallic copper nuclei, at low temperatures, become the first example of the nuclear spin-density waves.

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Extended Appearance-Potential Fine-Structure Analysis: Oxygen on Al(100)

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To measure O-Al separation at Al(100) surfaces disordered (low-energy electron-diffraction beams extinguished) by reaction with oxygen, the extended appearance-potential fine structure was analyzed above the threshold for electron-bombardment excitation of the O $1s$ core. Calculation shows that the outgoing electron has angular momentum $l = 0$, allowing simple Fourier inversion of the fine structure. The separation, 1.98 ± 0.05 Å, suggests that oxygen lies under the top layer, a result undetectable in extended-x-ray-absorption fine-structure measurements on thicker films.

The reaction of a (100) single-crystal aluminum surface with oxygen suppresses completely the low-energy electron-diffraction (LEED) pattern, once oxygen coverages exceed roughly a monolayer.[1] This loss of long-range order renders techniques such as LEED and ion backscattering incapable, even in principle, of determining the oxygen-metal distance. In a recent paper, however, Stöhr, Denley, and Perfetti[2] demonstrated that surface extended x-ray absorption

fine-structure (EXAFS) analysis, using mono-chromatized synchrotron radiation, can provide information on the oxygen-metal distance in aluminum oxide layers approximately 30 Å thick. We demonstrate that extended appearance-potential fine-structure (EAPFS) analysis can be used to obtain interatomic distances for much thinner layers. It is therefore well suited to studies of overlayers and is capable of extracting information inaccessible to probes requiring greater thickness. Moreover, EAPFS requires only an inexpensive electron source for excitation.

Fine structure in the excitation probability of core states by electron bombardment is observed to extend for hundreds of volts above the excitation thresholds or "appearance potentials."[3] It results from the interference of an outgoing state with backscattered components from neighboring atoms and is therefore analogous to EXAFS.[4] Accordingly, interatomic distances can be extracted by essentially the same Fourier inversion techniques. EAPFS has been used previously to de-termine nearest-neighbor distances in the surface region of polycrystalline vanadium[5] and other transition metals,[6] but this experiment is the first to illustrate its sensitivity to thin overlayers and to obtain a precise spacing.

The surface sensitivity is a consequence of the short mean free path for inelastic scattering of the incident electrons. Similar sensitivity has been achieved in EXAFS by use of the Auger yield to signal the creation of core holes for I/Ag(111).[7] That technique cannot be applied to oxygen adsorbates, since the apparent Auger yield is strongly modulated by core electrons photoemitted from the substrate.[2] The core-state photoelectron peaks are swept through the window of the electron analyzer as the incident wavelength is varied. To avoid this difficulty, Stöhr, Denley, and Perfetti[2] used a partial-yield technique,[8] in which the analyzer window is set in the inelastic tail of the spectrum. Thus, in order for an adsorption event to be monitored, it must occur sufficiently far inside the solid—roughly one inelastic mean free path—for the excited electron to scatter inelastically.

Electron excitation of a core state in metals is contrasted to x-ray absorption schematically in Fig. 1. In the x-ray case a single electron is ejected; its energy is the difference between the incident photon energy and the core binding energy. In the appearance-potential experiment, the
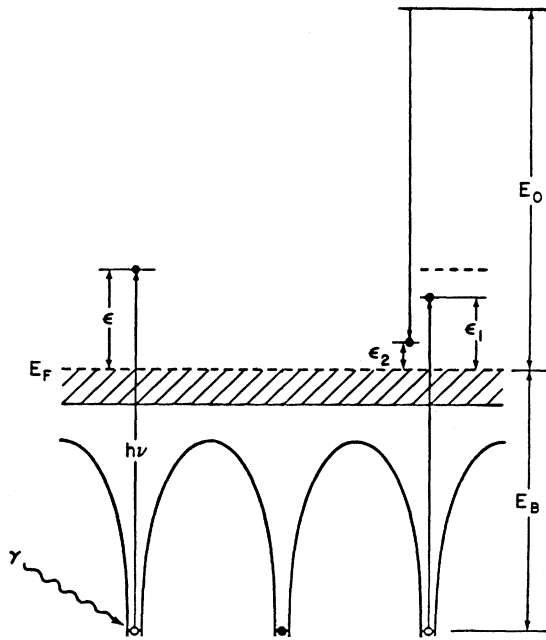

FIG. 1. Energy-level diagram comparing the excitation of a core level by a photon (left side) to excitation of the level by an electron (right side). Because both the incident electron and the ejected core electron must be accommodated in states above the Fermi level, the final state is a two-electron state. Differentiation with respect to E_0 selects the situation in which one electron is at E_F and the other carries the remaining energy (as in the x-ray case).
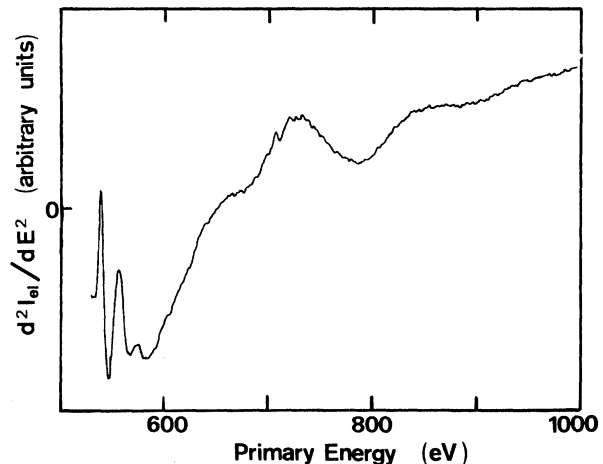

FIG. 2. Second derivative of the elastic yield of the oxidated Al(100) surface, plotted as a function of incident electron energy. The O 1s appearance-potential edge is at 533 eV (correcting for the emitter work function). Fine structure extends several hundred volts above the edge, but the periodic variations are masked considerably by the slowly-varying background.

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final-state energy is divided between two elec-
trons. However, differentiation of the yield with
respect to incident energy E_0 produces a signal
which strongly emphasizes the situation in which
one of the final-state electrons lies at the Fermi
energy or, more generally, the lowest unoccu-
pied state.[3,9,10] Thus, it is the first derivative of
the yield which is analogous to the undifferenti-
ated absorption spectrum in EXAFS.

Our measurements were obtained with conven-
tional hemispherical-grid LEED optics. We
measured the second derivative of the elastic
yield as a function of E_0. This yield is obtained
by biasing the retarding grids a few volts positive
with respect to the emitter of the electron gun.
At the thresholds for inelastic scattering from
core states, the elastic yield decreases.[11] Jach
and DiStefano[12] reported EAPFS in elastic yield
measurements, but analyzed to only about 100
eV above threshold. Substantial multiple scatter-
ing seriously complicates most of this region,
and may explain their failure to obtain reasonable
spacings. In our measurements, the second de-
rivative was obtained by superimposing a small
sinusoidal oscillation on the sample potential and
detecting at the second harmonic.

By operating the LEED optics as a retarding-
potential analyzer, we obtained the Auger-elec-
tron spectrum of the surface and thus could moni-
tor surface coverage. A clear aluminum surface
was obtained by argon-ion bombardment and an-
nealing. After exposure to 120 L of oxygen [1 L
(langmuir) = 10^{-6} Torr sec], the LEED beams
were completely extinguished; this exposure cor-
responds to about 1.5 equivalent monolayers of
oxygen.[13] We ramped the incident electron ener-
gy range from 500 to 1000 eV. Only the oxygen
1s edge at 533 eV lies (Fig. 2) in this range. The
aluminum 1s is at 1540 eV and the Al 2p occurs
at 73 eV. Some fine structure from the Al 2p
presumably extends beyond the O 1s, but is too
weak to interfere with measurements of the oxy-
gen spectrum.

Variations in the elastic yield resulting from
diffraction of the incident electron beam often ob-
scure appearance-potential features in the energy
range up to perhaps 600 eV. In the case of the Al
(100) surface, however, the oxygen-induced loss
of long-range order suppressed these diffraction
variations almost completely. The remaining
slowly varying (compared with the fine structure)
background variations were removed from the da-
ta by first subtracting a least-squares-fit cubic
polynomial from the data to remove very coarse

variations[5] and subsequently applying a high-pass
digital filter[6] (Fig. 3).

A serious question remains. The phase shift
appropriate to the central (absorbing) atom and
the surrounding backscattering atoms differs for
the various angular momentum components of the
outgoing electron. For a K electron the dipole
selection rule indicates that x-ray excitation
must place it in an $l = 1$ state. For electron exci-
tation, an explicit calculation is required to find
the relative coupling to each partial wave of the
higher-energy final-state electron. To make this
determination, we obtained the core state to be
excited from a Hartree-Fock-Slater calculation.[14]
For the other three wave functions, we used or-
thogonalized plane waves decomposed by angular
momentum about the central atom. We then cal-
culated Coulomb-potential matrix elements.[10]
The results[15] show that (1) orthogonalization to
the core state is crucial; (2) matrix elements are
smooth, slowly varying functions of energy; and
most importantly, (3) the matrix element coup-
ling to the $l = 0$ higher-energy outgoing final state
is overwhelmingly dominant, so that the oscilla-
tory part of the transition rate contains a single
term[16] of the form $F \sin[2kR_1 + \varphi(k)]/k^3 R_j^2$, where
k is the wave vector of the outgoing state, F is a
slowly varying envelope function dominated by
Debye-Waller effects, R_1 is the distance to the
nearest shell of surrounding atoms, and φ is the
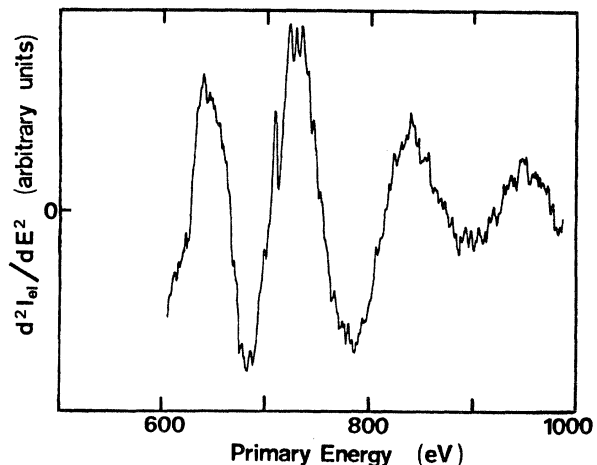k-dependent phase shift.

FIG. 3. Extended fine structure associated with the
O 1s edge plotted in Fig. 2. The first 70 eV of data
above the edge have been excluded to avoid complica-
tions related to multiple scattering. A polynomial
subtraction and digital filtering technique have been
applied to suppress the slowly varying background.

The $l = 0$ combined phase shift for the central (oxygen) and backscattering (aluminum) atoms was calculated with self-consistent nonrelativistic Hartree-Fock-Slater ($\alpha = \frac{2}{3}$) atomic potentials overlapped in the appropriate configuration. These results agree with calculations by Teo and Lee.[17] Using the threshold for the oxygen $1s$ excitation as the momentum zero,[18] we took an optical Fourier transform of the data from 70 to 450 eV above the edge, weighted by the cube of wave vector in standard EXAFS fashion.[19]

The magnitude of the transform is plotted as a function of interatomic spacing in Fig. 4. A single dominant peak, presumably corresponding to the nearest-neighbor oxygen-aluminum distance, is located at 1.98 ± 0.05 Å. The value corresponds to the longer of the two Al-O spacings in bulk Al_2O_3, viz. 1.97 Å (the shorter is 1.86 Å).[20] Our value is consistent with the number 2.02 Å proposed in self-consistent-field $X\alpha$ scattered-wave calculations of low-coverage O on Al(100).[21] Since the longer Al-O spacing in Al_2O_3 corresponds to the oxygens lying between two Al atoms (rather than between Al and a vacancy),[22] our result supports the idea that the adatoms go under rather than on top of the first Al layer. This picture had been advanced earlier on the basis of the decrease in work function with oxidation[23] and on secondary-ion mass spectrometer results.[24] The EXAFS result for the thick oxide layer was 1.91 Å,[2] the average of the two bulk spacings,

and thus was insensitive to this surface-related feature. The indicated error bars are a conservative estimate. The peak is shifted by less than 0.03 Å over a wide range of filter parameters.[25]

Our technique relied heavily on the disordering of the surface by oxygen to suppress diffraction variations in the elastic yield. Such variations are absent in soft–x-ray appearance-potential spectra. EAPFS studies of low-energy edges will therefore have to rely on the soft–x-ray method in most cases. The present results, however, demonstrate the feasibility of the EAPFS technique for the study of overlayers. The use of electron-bombardment excitation makes the technique available to essentially any surface physics laboratory.
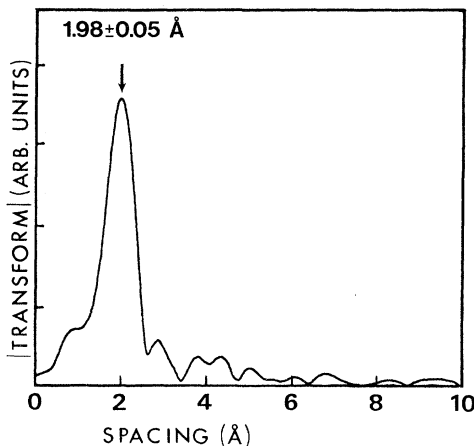
FIG. 4. Optical Fourier transform of the data in Fig. 3, including calculated phase shifts for an O $1s$ core and Al backscatterers. The single peak at $R = 1.98 \pm 0.05$ Å is taken as the O-Al spacing in this thin aluminum oxide layer, suggesting that the O lies under rather than over the top layer of Al. See text.

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ERRATUM

OPTICAL ISOMER SHIFT FOR THE SPONTANEOUS-FISSION ISOMER ^{240}Am^m. C. E. Bemis, Jr., J. R. Beene, J. P. Young, and S. D. Kramer [Phys. Rev. Lett. **43**, 1854 (1979)].

In line four of the abstract change 6407.2 ± 0.2 Å to 6407.7 ± 0.2 Å. Page 1855, left column, line 26 change "opitcal" to "optical." Page 1855, right column, line 26 change "rejected" to "ejected." Page 1856, left column, line 12 change "with use of" to "with the use of." Page 1857, left column, line 30 change "isotope of isomer" to "isotope or isomer."

In addition, two very recent review articles have appeared which deal with laser applications in nuclear physics and the optical spectroscopy of short-lived nuclides: (1) D. E. Murnick and M. S. Feld, Annu. Rev. Nucl. Part. Sci. **29**, 411 (1979), and (2) H. J. Kluge, in *Progress in Atomic Spectroscopy B,* edited by W. Hanle and H. Kleinpoppen (Plenum, New York, 1979), p. 727. The interested reader is directed to these for additional background information.