

Low-energy electron and positron diffraction measurements and analysis on Cu(100)

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The results of an experimental study and quantitative analysis of the intensity versus energy (I - V) curves are reported for low-energy electron diffraction and low-energy positron diffraction (LEPD) with a brightness-enhanced electrostatically focused positron beam. In a close comparative study, the incident electrons and positrons scattered at a large incident polar angle ($\theta \geq 50^\circ$) with respect to the surface normal off clean Cu(100) and the I - V spectra from six and seven diffraction beams were taken with electrons and positrons, respectively. The analysis of the experimental data from the electron studies indicates first- and second-layer relaxation that is consistent with earlier results. Use of the structure derived from the electron studies, analysis of the I - V curves from the LEPD studies suggests that the attenuation for positrons is greater than the value for electrons over the energy range 50–400 eV, possibly as a result of the enhanced electron-image cloud surrounding the positron. The real part of the inner potential is 0 eV for positrons compared with 11 eV for electrons, in rough agreement with predictions. Further, the best agreement between experiment and calculation for LEPD I - V curve analysis tends to favor the potential formed by changing of the sign of the Coulomb term (relative to electrons), eliminating the exchange, and retaining the correlation term.

I. INTRODUCTION

Low-energy electron diffraction (LEED) is one of several techniques that determine the position of atoms near the surface region of a bulk crystal. The long-range order for atoms situated near the surface region can be determined through LEED by analyzing the geometrical pattern of the diffraction spots. In addition, the relative positions of the surface atoms can be deduced by comparing experimental and theoretical intensity versus incident beam energy (or “ I - V curves”) for a set of diffraction beams. The appropriate structural and nonstructural parameters in the model are then varied so that the predicted I - V spectra agree with the experimental results. Such a scheme relies on knowledge of many-particle potentials such as the energies arising from electron exchange and correlation.

For electrons, studies of many surfaces of metals and semiconductors show that the self-consistent potentials used in band structure calculations are adequate¹ to describe even very small structures in the experimental LEED data. There is some question whether this remains true for transition elements.¹ The calculated spectra of tungsten (100) (Ref. 2) and molybdenum (100) (Ref. 3) are also more dependent on the details of the potential than other materials. However, many calculations for lighter materials show practically no disagreement with respect to experiment from¹ 50 to 200 eV, or even 300 eV of elec-

tron energy in spite of the fact that the band-structure potential used for the study is based on Kohn-Sham theory and the local-density approximation which are really only justifiable for ground-state properties. Furthermore these self-consistent potentials produce noticeably better spectra than the superpositions of atomic potentials which have been used in band calculations in the past. This shows that the spectra probes the details of the potential and not, for example, just the core electron density.

To improve the accuracy and precision of surface structure analysis via I - V curves, it has been suggested that low-energy positron diffraction (LEPD) may complement conventional LEED.^{4–6} Aside from simply extending the data base, LEPD should provide complementary information for LEED. For instance, the exchange interaction is absent and the real part of the inner potential is thought to be known in principle for positrons.⁶ It has been suggested therefore that uncertainties are reduced in the experimental and theoretical LEPD I - V curve comparisons.

In two seminal studies, Rosenberg *et al.*⁴ and Weiss *et al.*⁵ improved the positron-beam parameters (although their apparatus did not match the beam profiles or the beam width and beam angle of conventional LEED systems) and measured the first specular and nonspecular diffraction I - V curves from Cu(111) and Cu(100). To achieve reasonable agreement in the comparison of experimental and theoretical I - V curves, it was necessary to perform an average over an angle of $\pm 2^\circ$ due to beam diver-

gence and the wide angular acceptance of their detector. The comparison was limited to the 00 and 10 diffraction beams. Mills and Platzman,⁷ Mills and Crane,⁸ and Cook *et al.*⁹ have also observed the specular I - V curves of positrons off various surfaces.

Frieze *et al.*¹⁰ constructed the first brightness-enhanced electrostatically focused (BEEF) positron beam with approximately a beam diameter and angular divergence of 1 mm and 1° , respectively, over the energy range 50–500 eV and that apparatus was used in the present study. Such characteristics are similar to a conventional LEED beam although the positron beam had a much lower intensity. A channel electron multiplier array (CEMA) coupled to a resistive anode detected the low current from the scattered beams. In addition, the CEMA permitted the simultaneous collection of I - V curves from several diffraction beams.

Until recently, it has not been experimentally feasible to conduct adequate tests of LEED and LEPD due to the poor phase characteristics of previous positron beams. This paper discusses the use of a positron BEEF beam and quantitative analysis for LEPD and LEED at large polar angles for the Cu(100) surface. The Cu(100) surface is well studied with generally accepted structural and non-structural parameters. This surface was therefore used as a standard to demonstrate that LEED and LEPD can be employed as a quantitative surface probe. In addition, vacuum contaminants have a low sticking probability on Cu(100) at room temperature. The electron and positron beams hit the sample at non-normal incidence and seven and six nondegenerate spectra were collected from the positron and electron beams, respectively.

This paper discusses the essential aspects of the experiment and analysis that are relevant to this non-normal incident-beam LEPD and LEED study. In Sec. II, we present a description of the apparatus, data collection, and experimental procedure for determining the polar and azimuthal angles. Section III discusses the analysis and results of the LEED and LEPD data and how the analysis confirmed our experimentally derived incident beam angles. The last section summarizes the paper and discusses future prospects and improvements in the apparatus.

II. EXPERIMENT

As had been described in an earlier publication,¹⁰ this experimental chamber was equipped with a brightness-enhanced electrostatically focused (BEEF) positron beam¹¹ system and a low-current ($\approx 10^4$ – 10^5 s⁻¹) electron gun and will briefly be described. In addition to the positron and electron-beam system, this chamber has a reverse-view low-energy electron-diffraction (LEED) system, a retarding field Auger electron spectrometer (AES), an ion-sputtering gun, a residual gas analyzer (RGA) and an electron bombardment gun (Fig. 1).

The positron-beam system, shown schematically in Fig. 2, consists of a ^{58}Co positron source, a moderator, two remoderators, and three electrostatical lens sections. A reflection moderator of W(111) coated with oxygen was used to produce low-energy positrons from a $1 \times 3\text{-mm}^2$ ^{58}Co (≈ 500 mCi) positron source electroplated onto a

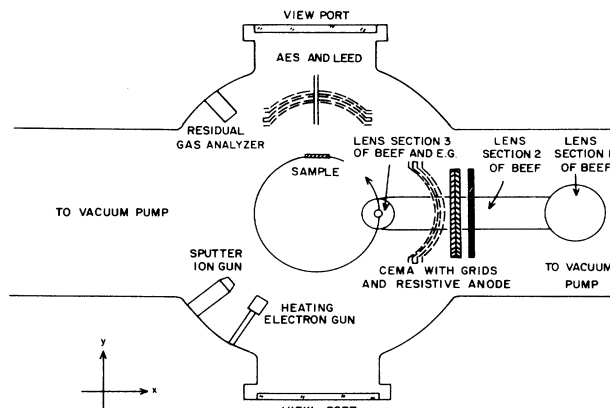


FIG. 1. Apparatus showing target chamber with CEMA, sample, positron BEEF beam, sample preparation probes, sample analyzers, and "X-Y" axis.

tungsten single crystal plated with copper. The reemitted positrons from the moderator are accelerated to 2 kV and projected about 2 m to extract the beam from the background of high-energy β^+ particles and γ radiation from the ^{58}Co source. Due to the electric field distortion in the source-moderator region, the first lens section only produced a 3-mm-diam positron beam on the first remoderator (RM1). The first remoderator is a single crystal of Ni(111) coated with adsorbed Co. By adding a coating to the Ni(111) crystal, the total slow-positron yield from RM1 is enhanced relative to the clean crystal due to the increase in the negative positron work function and thus the positron yield. The major improvement of the phase-space characteristics of the positron is, however, achieved in lens section 2. In this section, the two remoderators (RM1 and RM2) and the second lens section are well shielded by soft iron and μ metal, respectively. The positron beam from the RM1 is successively accelerated by the first lens set triplet to provide a nearly parallel beam passing through two pairs of deflection plates and then

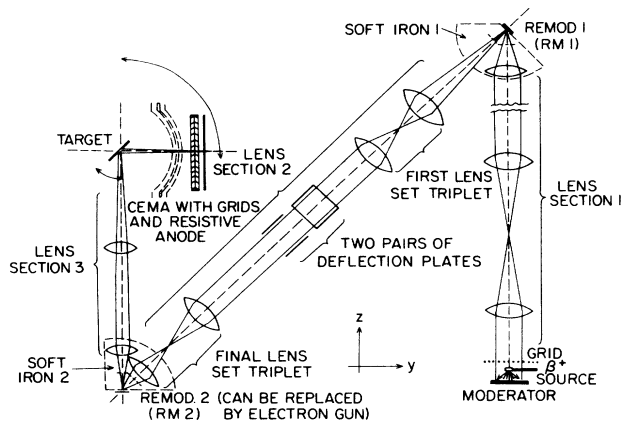


FIG. 2. View of positron BEEF beam, target, and CEMA detector. 90° rotation from Fig. 1.

strongly focused by the final triplet lens set onto the second remoderator (*RM2*) of a single crystal W(110) coated with oxygen. The positron beam at *RM2* had a diameter of approximately 0.2 mm. The third lens section delivers the doubly remoderated beam to the target with about 1-mm diameter, $<1^\circ$ opening angle, and the estimated energy width of 0.07 eV at a typical beam energy of 200 eV. An electron gun can be rotated into the position of *RM2* and produce an electron beam which, like the positron beam, passes through lens section 3 and hits the sample. The energy of the positron or electron beam can be tuned from 20–500 eV by biasing *RM2* or the electron gun to the desired voltage. The energy of the positron beam is determined by this voltage plus the work function of positron on W(110) with chemisorbed oxygen or *RM2*. The overall conversion efficiency of the positron beam in this system as delivered to the target is $\approx 2 \times 10^{-5}$ or in other words, with a 500-mCi source of ^{58}Co , 5×10^4 positron per sec at energy of 500 eV can be directed at the target.

Positrons or electrons scattered from the sample were detected with a 7-cm-diameter channel electron multiplier array (CEMA) coupled to a resistive anode mounted behind the CEMA¹² and four grids (80% transmission each) placed in front of the detector. This detector provides two-dimensional information of scattered positrons or electrons and has a spatial resolution of <0.5 mm full width half maximum (FWHM). To minimize the background unrelated to the incident beam, such as “hot spots” (small regions on the CEMA which generate anomalously high current, possibly due to the presence of fiberglass or dust on the plate) the grids were modulated. That is, the data were set to the difference in detected counts with the grids fixed at a value of 10 V below the incident-particle energy (accepting elastically scattered events), minus the detected counts with the grids set to a voltage 10 V above the particle energy (suppressing the incident beam).

The two-dimensional data were stored and partially analyzed with an on-line computer. Before the current from the diffracted beams was measured, the target was retracted from the particle-target interaction region, the incident beam intensity at each energy was collected, stored, and subsequently used to “normalize” the diffraction beam current. In addition, uniform spatial-detection efficiency was checked by shining light on small areas on the channel plate-resistive anode assembly and observing the detected counts as the position of the incident light beam was varied. During an *I-V* measurement, several diffraction beams hit the CEMA detector at the same time and could be collected in a single run. It was therefore necessary to only collect counts from the diffraction beams within appropriate spatial windows as shown in Fig. 3. For fixed incident-beam–target orientation, all diffraction beams (except the specular) move as the incident beam energy is varied. To generate an *I-V* curve for nonspecular spots over a large energy range the data collection region was divided into several overlapping rectangular data windows. Individual *I-V* curves were taken from each of the small rectangular regions. These curves were subsequently “welded” together to form a single

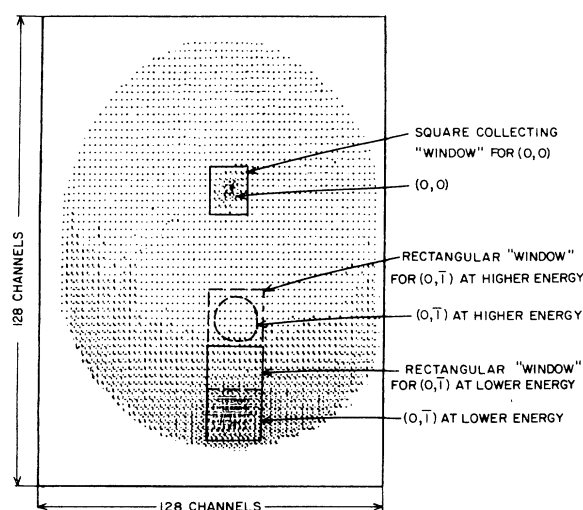


FIG. 3. 2D intensity plot of 00 and $0\bar{1}$ beam at low energy. Data collection windows for the diffraction spots also shown.

curve for the total energy region. That is, the normalized *I-V* curve values within one spatial-energy window were linearly rescaled so as to match the value of the normalized *I-V* curve of the adjacent window within the overlap region. The specular beam does not move and therefore was collected within a single window. Using this method, the collection of some background within a window was unavoidable. The *I-V* curve, however, was dominated by the diffraction beam and not the background which tended to vary slowly with incident-beam energy. (The background is proportional to the counts inside spatial windows located far from the windows associated with the diffracted beams and is a slowly increasing function of incident beam energy). The shape of the *I-V* curves were not strongly affected by the geometry of the collection windows.

During the measurement of the *I-V* curve, the energy was swept from lowest to highest energy in 2- and 3-eV steps for electrons and positrons, respectively. The counts collected from the “window” for each energy were stored in the computer. The dwell time for each energy step was about 50 and 200 s for electrons and positrons respectively. The *I-V* curve data were collected by making about 3–4 such energy sweeps to reduce the effect of beam drifts and summed. The total collecting time for each energy was therefore about 200 s for electrons and 500–800 s for positrons. The statistical error in the normalized diffracted beam intensity was less than 5% at each energy.

The whole CEMA analyzer is mounted on a rotating cradle which allows it to be turned through 90° from the vertical position shown in Fig. 1 to a horizontal position. The incident-beam polar angle (θ) can be varied from 45° to 90° with respect to the target surface normal by rotating the sample holder. Unfortunately, the sample can only rotate in the θ direction. The azimuthal angle (ϕ), or the angle between the component of the incident beam

parallel to the sample surface and with respect to the sample unit cell, cannot be adjusted after the sample is installed in the vacuum system. We independently measured the direction of the incident beam with respect to the sample holder, the azimuthal angle direction to a reference line, and the orientation of the sample surface to the same reference.

To measure the polar angle θ of the incident beam, we moved the sample holder in direction X (Fig. 2) and used a beam blocker mounted on the sample holder to partially and eventually totally prevent the beam from striking the CEMA. The CEMA in this case simply measured the beam intensity. A plot of the CEMA counts versus sample (or blocker) position is the integrated profile of the beam. In other words, the derivative of this curve yields a Gaussian distribution whose width and peak position are the beam width and beam center, respectively. This measurement was repeated for different vertical positions (or Z) of the blocker and the orientation of the beam with respect to the sample holder could be measured by comparing the peak positions for two different values of Z . The sample was then rotated and photographs were taken of the sample from a direction parallel to the surface of the sample to get the angle between the sample surface and the sample holder. We took a multiple-exposure picture with a rigidly mounted camera from the Y direction to find the angle between a line delineated by sample holder due to movement in direction X , and the sample surface. From these results, θ was measured to within an error of about $\pm 0.8^\circ$.

Similarly, in the ϕ measurement, we moved the sample holder in direction Y (Fig. 1) to two different heights and used the blocker and channel plate to get the orientation of the beam with respect to the sample holder. In addition we took pictures of a symmetrical LEED pattern from a reverse view LEED apparatus to get the orientation of the unit cell of the crystal relative to the shaft. We then took a multiple-exposure picture along the X direction to find the angle between the line delineated by the shaft from movement in direction Y and the sample. From these results, ϕ was measured to within an error of about $\pm 0.8^\circ$.

Stray electric and magnetic fields in the beam-target region affect the trajectory of electrons and positrons and alter the incident beam angle of the particles as they collide with the target. The change in the particle path due to residual fields is beam energy dependent and can introduce an error into the I - V curves. To eliminate these fields, three mutually perpendicular Helmholtz coils nulled the stray fields. A number of methods were used to detect the presence of these fields and check that the strong fields were eliminated. Using the CEMA as an accurate beam-angle detector ($\leq 0.1^\circ$) in separate experiments, the positions of the specular and the incident beam (target removed from the interaction region) were measured as a function of particle energy. Finally, we measured the center of the integrated beam profile (using the beam blocker and CEMA as a current detector) as a function of beam energy. Over the energy ranges (50–450 eV) of I - V curves, the fields were balanced so that the incident-beam–target angular variation was $\leq 0.3^\circ$. The

same magnetic field orientation and magnitude was required to null the fields for both the LEPD and LEED I - V curve measurements. During the measurement of a complete set of I - V curves, the voltages on the lens and deflection plates were fixed and the position of the moderators or electron gun was not altered to avoid changes in beam orientation.

The Cu(100) sample is about 25 mm in diameter, oriented to within 1° and was mechanically and chemically polished. It was then heated in a hydrogen and argon atmosphere at 600°C for 48 h to remove any sulfur remaining in the bulk crystal. This sample had a low dislocation density of 10^{-4} cm^{-2} and was 99.999% chemically pure. The sample was cleaned by sputtering with Argon ions at 0.5–1 keV for 5–10 min, subsequently annealed at 600°C for 5 min, and slowly cooled. To find a rough quantitative value for adsorbate coverage, the AES spectra from a saturated overlayer of oxygen in the $c(2\times 2)$ superstructure on Ni(100) was used as a standard and compared to our AES spectra. After accounting for differing Auger sensitivities at various energies, the surface contamination was found to be less than 0.1 monolayers of oxygen, carbon, and sulfur. Large diffraction peaks from the copper were observed in the AES spectra.¹³

III. RESULTS AND ANALYSIS

LEED analysis

The LEED data were collected on the same sample and in the same configuration as the LEPD data in order to test the experimental geometry and to allow for refinement of the structural parameters of the Cu(100) surface. The latter requirement was particularly important in order to eliminate or at least reduce the uncertainties introduced by the structural parameters in the forthcoming tests of the LEPD experiments. The present experimental geometry was rather unusual for LEED, which is mostly done with angles of incidence at or near normal ($\theta=0^\circ$): in the present experiments normal incidence was not attainable and the angle θ was mostly larger than about 50° . In addition, the azimuthal angle ϕ (for a definition of these angles in LEED crystallography see, e.g., Ref. 14) could not be varied *in situ* and in the present experiments was close but never along symmetrical directions. This rather uncommon situation made it desirable, if not necessary, to check the experimental values of θ and ϕ with intensity calculations. Thus, the procedure adopted for the LEED analysis was the following. First, with the nominal values of θ and ϕ as determined experimentally, the fit of theory to experiment was optimized by varying independently the values of the first and the second interlayer spacing (d_{12} and d_{23} , respectively) of the Cu(100) sample. With the best values of d_{12} and d_{23} so determined, the fit of theory to experiment was further optimized by varying θ and ϕ around the experimentally determined values. Finally, with the best values of θ and ϕ thus determined a second refinement of d_{12} and d_{23} was carried out. In all tests, the fit between theory and

experiment was quantified by means of the Zanazzi-Jona r factor.¹⁵

The Zanazzi-Jona r factor is a measure of the goodness of fit between the experiment and theory and is often used for I - V curves. The r factor includes the first and second derivatives of I - V curves with respect to the incident-beam energy and is therefore suitable for smooth data. Unlike conventional LEED detectors, the CEMA coupled to a resistive anode measures pulses and can only accept low count rates (<20 kHz) in order to avoid saturation. The intensity of the I - V data in this experiment therefore has statistical errors ($<5\%$ error at each point). To use the Zanazzi-Jona r factor, the experimental data were smoothed to minimize the effects of the statistical error and avoid artificially large r factors (due to an excessively large first and second derivatives) without eliminating the real maxima and minima from the experiment.

The LEED intensity calculations were done with the computer program CHANGE described elsewhere.¹⁶ The Cu potential was obtained from the Moruzzi, Janak, and Williams compilation¹⁷ and 8 phase shifts and 45 beams were used to describe the wave function. The inner potential was chosen as $V_i = V_0 - i\beta$, with V_0 independent of energy and initially fixed at -10 eV, the final value to be determined from the intensity analysis, and β a function of energy as $\beta = 0.85E^{1/3}$ eV (see Refs. 18 and 19). The mean vibrational amplitude was taken as 0.15 Å for both bulk and surface atoms.

The experimental data base consisted of six spectra: 00, $0\bar{1}$, 10 , $\bar{1}0$, $\bar{1}\bar{1}$, and $1\bar{1}$, the first 2 extending from 40 to 400 eV, the rest from 150 to 380 eV for a total energy interval of 1638 eV. The angles of incidence were measured as $\theta = 52.0^\circ \pm 0.8^\circ$ and $\phi = 91.7^\circ \pm 0.8^\circ$.

The present work was initially facilitated by the fact that a multilayer analysis of Cu(100) had previously been done and reported in the literature by Davis and Noonan.¹⁹ The results of that analysis was that d_{12} is contracted by 1.1% (or 0.02 Å) d_{23} is expanded by 1.7% (or 0.03 Å) and d_{34} is expanded between 1% and 2% with respect to the bulk value 1.807 Å, and the real part of the inner potential is -10 eV. Thus, in the present work the first round of calculations involved variations of d_{12} from the bulk value to a value contracted by 4.4% in steps of 1.1% ($=0.02$ Å), and independent variations of d_{23} , also in steps of 1.1%, from a contraction of 1.1% to an expansion of 5.5%. The minimum r factor value in this range of the d_{12}, d_{23} plane was 0.126 for d_{12} contracted by 3.4% and d_{23} expanded by 2.0%.

The second round of calculations involved variations of θ from 48° to 52° , and of ϕ from 89° to 95° in steps of 1° , the values of d_{12} and d_{23} being kept fixed at the level determined in the first round. The minimum r -factor value in the range of the θ, ϕ plane defined above was 0.072 for $\theta = 51.3^\circ$ and $\phi = 93.3^\circ$ (versus the experimental values of $\theta = 52.0^\circ \pm 0.8^\circ$, $\phi = 91.7^\circ \pm 0.8^\circ$).

The third and final round kept θ and ϕ fixed at these values and varied d_{12} and d_{23} again as in the first round. The minimum r factor dropped to 0.064 for d_{12} contracted by 2.1%, d_{23} expanded by 0.45% and $V_0 = -11$ eV. A comparison of the predicted and experimental values for LEED I - V curves are shown in Figs. 4 and 5.

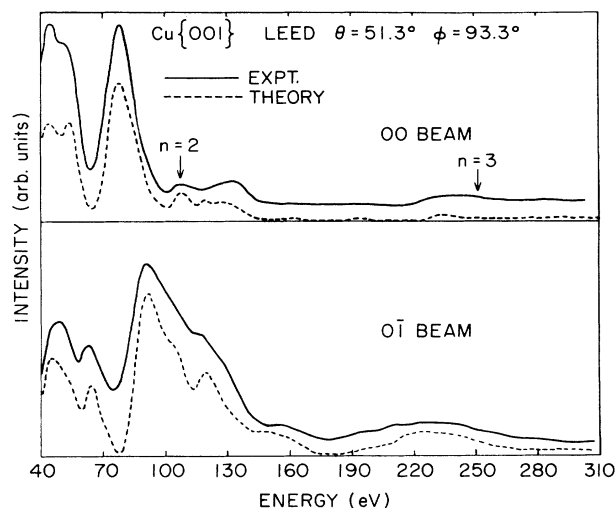


FIG. 4. Experimental and calculated normalized LEED I - V curves for 00 and $0\bar{1}$ beams. Arbitrary units used in vertical scale. Experimental and calculated curves are shifted to aid visual comparison. Arrows in 00 curve denote $n=2,3$ Bragg maxima.

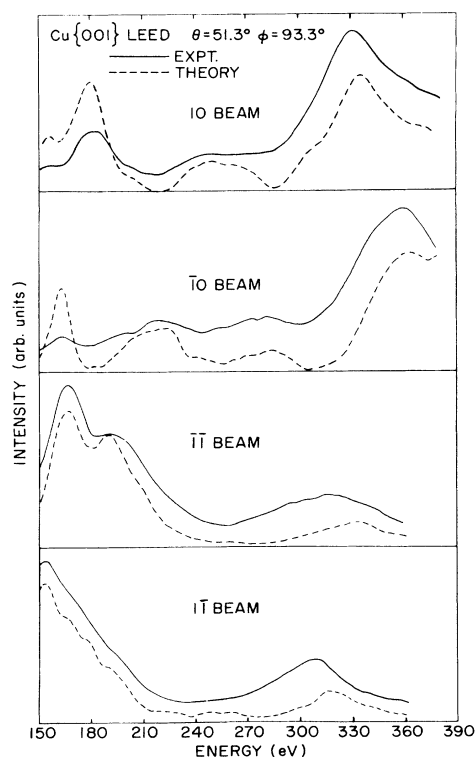


FIG. 5. Same as Fig. 4; 10 , $\bar{1}0$, $\bar{1}\bar{1}$, and $1\bar{1}$ beams.

Two comments about these results may be appropriate. The first is that comparison of the structural parameters with the literature values, while perhaps not as satisfactory as one would like, reveals agreement within our estimated error of about 0.03 Å or 1.7% (See, e.g., Shih *et al.*, Ref. 20). The second comment is that the data base used in the present analysis, six spectra over a total energy range of about 1600 eV, is only marginal for the purpose of high reliability in the structural parameters, and in view of the usually large non-normal angles of incidence.

In conclusion, we summarize below, for the convenience of the reader, the results of the present LEED analysis: bulk interlayer spacing 1.807 Å, $d_{12} = 1.769 \pm 0.03$ Å (contracted 2.1%), $d_{23} = 1.815 \pm 0.03$ Å (expanded 0.45%), $V_0 = -11 \pm 2$ eV, $\bar{r}_r = 0.064$ for six spectra ($\Delta E = 1638$ eV).

LEPD analysis

The LEPD analysis was initiated on the assumption that the structural parameters d_{12} and d_{23} of the Cu(100) sample were known from the LEED study (see above), and that the quantities to be determined were the non-structural parameters $V_i = V_0 - i\beta$ (the inner potential) and the "best" positron potential for LEPD work. In addition, the incidence angles θ and ϕ , although determined experimentally, were to be optimized by the intensity analysis. The criterion for all these determinations was to be the quality of fit between calculated and experimental spectra as determined both visually and by r -factor analysis, just as in the LEED work.

The data base consisted of seven spectra, 00, $0\bar{1}$, 10, $\bar{1}0$, $1\bar{1}$, $\bar{1}\bar{1}$, and $0\bar{2}$ for a total energy range of 1485 eV at $\theta = 52.5^\circ$ and $\phi = 91.7^\circ$. The first few intensity calculations revealed that the real part V_0 of the inner potential had to be set equal to approximately 0 and the absolute value of the imaginary part β had to be larger than that used in the LEED calculations. Minimization of the r factor turned out not to be useful in determining the best value of β because the r -factor values kept becoming unrealistically small with increasing values of $|\beta|$. A procedure such as that followed by Demuth *et al.*¹⁸ for the determination of the energy dependence of β (i.e., the measurement of the halfwidths of intensity peaks) was not possible because all the intensity maxima occurring in the energy and angle ranges investigated here appeared to consist of several adjacent diffraction peaks. Hence, we resorted to an approximate determination of $|\beta|$ by choosing the smallest such value (independent of energy) that would provide acceptable visual agreement between theoretical and experimental spectra. This value was set at $|\beta| = 6$ eV.

The phase shifts needed for the intensity calculation were obtained from three different potentials. The exchange part of the exchange-correlation term does not exist for positrons. In addition, the correlation energy associated with the interaction of positrons with core electrons is expected to be small²¹ due to the repulsion between the positrons and the ion cores. Potential P_C was the same potential that produced satisfactory agreement with experiment in earlier LEPD calculations²² (this potential

was denoted P_1 in Ref. 18) and was constructed from the Moruzzi-Janak-Williams¹⁷ (MJW) Cu potential by changing the sign of the Coulomb contribution, eliminating the exchange and retaining the correlation term. The correlation term in this case was one fourth the size of the exchange part which was dropped. Potential P_{EC} was constructed from the MJW potential merely by changing the sign of the Coulomb part but keeping the exchange and correlation terms as applicable to electrons, while potential P_{NEC} had also the sign of the Coulomb contribution changed but no exchange or correlation included. The initial calculations, aimed at the optimization of the incidence angles θ and ϕ and of the complex inner potential $V_0 - i\beta$, were done with potential P_C , which is expected to produce the best fit between the theory and the data.

With potential P_C and $\beta = 6$ eV calculations done with the experimental values $\theta = 52.5^\circ \pm 0.8^\circ$, $\phi = 91.7^\circ \pm 0.8^\circ$ and with the structural parameters determined with LEED (see above) yielded a minimum r factor of 0.061 for $V_0 = 0$. A series of calculations involving variations of both θ and ϕ in steps of 1° around the experimental values revealed a minimum r factor of 0.049 for $V_0 = +2$ eV at $\theta = 52.5^\circ$ and $\phi = 92.7^\circ$. These values of the incidence angles were henceforth assumed to be correct and used in all subsequent calculations.

Visual comparison of the results obtained with the three potentials P_C , P_{NEC} , and P_{EC} are shown in Figs. 6–8. We note that the agreement with experiment provided by any of the three potentials is satisfactory and the differences among the curves produced by them are small yet visually detectable. However, a decision as to which of the three is best in terms of agreement with experiment is

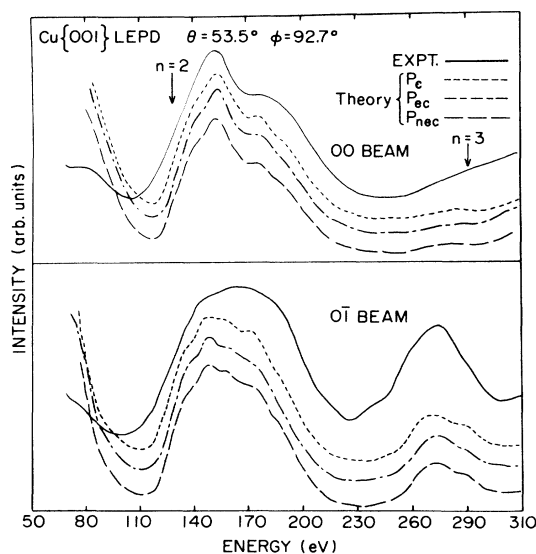
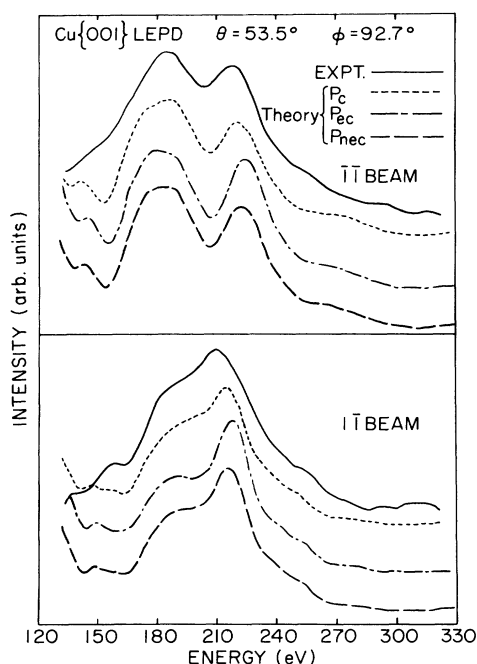
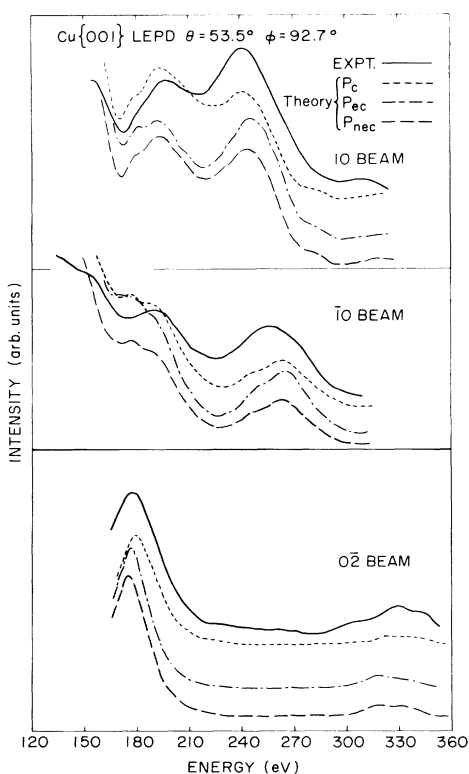


FIG. 6. Experimental and calculated normalized LEPD I - V curves for 00 and $0\bar{1}$ beams. Arbitrary units used in vertical scale. Experimental and calculated curves are shifted to aid visual comparison. Arrows in 00 curve denote $n=2,3$ Bragg maxima. Theory curves with $\frac{1}{6}$ exchange (P_C), full exchange (P_{EC}), and no exchange (P_{NEC}).

FIG. 7. Same as Fig. 6; $1\bar{1}$ and $1\bar{i}$ beams.FIG. 8. Same as Fig. 6; 10 , $10\bar{0}$, and $02\bar{0}$ beams.

difficult to make. The results for the three potentials are summarized thus ($V_i = V_0 - \beta i$): $\bar{r}_r = 0.049$, $V_i = 2 - 6i$ eV for potential P_C , $\bar{r}_r = 0.058$, $V_i = -1 - 6i$ eV for potential P_{EC} , $\bar{r}_r = 0.055$, $V_i = 0 - 6i$ eV for potential P_{NEC} . The differences in r -factor values are small. In rigorous terms, potential P_C has a slight edge over the others, but the differences are too small to allow confidence conclusions of general validity. Physical insight suggests that the best potential would have an intermediate value between P_C and P_{NEC} .

IV. DISCUSSION

The present study of LEPD and LEED has substantial improvements in the experimental technique and is, as a consequence, the first among the three such studies reported^{4,5,22} to use a data base of seven nondegenerate beams for comparison with the calculations. The lack of normal-incidence capabilities is unfortunate since normal-incidence data would complement data obtained here but the expanded data base makes a meaningful comparison between LEPD and LEED more reliable than it had been heretofore.

The experimental data for electrons was compared to the model used in the CHANGE program after accounting for the large polar and azimuthal angles. The lowest Zanazzi-Jona r factor (a measure of the best fit to the model) indicated surface relaxation of $d_{12} = -2.1 \pm 1.7\%$, $d_{23} = +0.45 \pm 1.7\%$ and the real part of the inner potential $V_0 = -11 \pm 2$ eV. These values agree with previous data¹⁹ obtained with normal incidence results within our stated error. Using the structure from our electron data, the positron data were analyzed and the best fit gave a value for the real part of the inner potential of $V_0 = 0 \pm 2$ eV, consistent with the suggestion of Read and Lowy⁶ that this parameter for positrons should equal the positron work function and with the results of Weiss *et al.*⁵ The apparent work function for electrons at higher energies does not equal the deduced value from the real part of the inner potential because the correlation contribution to the energy changes with the incident energy. This should also happen for positrons, so the measured shift V_0 need not necessarily agree with what is expected from the work function. The standard expression for the energy E_n of the n th order specular Bragg peak off a Cu(100) crystal is

$$E_n = \frac{11.5n^2}{\cos^2\theta} + V_0 \quad (\text{in eV}).$$

In Figs. 4 and 7, the position of the predicted second- and third-order Bragg peaks for electron and positrons are denoted by arrows in the specular I - V curves. The shift in energy of the experimental and calculated peaks with respect to the predicted Bragg positions, illustrates the importance of using the dynamical picture to describe the behavior of low-energy positrons and electrons, although the simple kinematic picture seems to be valid for electrons and positrons in NaF and LiF.⁸ The imaginary part of the inner potential or attenuation (a measure of inelastic processes) $\beta = 6$ eV was larger than the value for electrons (4 eV). The inelastic processes for positrons

broadened the peaks in the I - V curves and made more difficult a definitive r -factor analysis of the positron data and tests of the effects of exchange and correlation.

The present work finds that the damping or imaginary part of the inner potential for 50- to 450-eV positions is larger than its value for electrons at these large incident angles. The parameter β is a measure of the strength of the inelastic scattering processes and is generally attributed to electron-hole and plasmon production. Theoretical calculations^{23,24} suggest, however, that the production of these excitations by positrons and electrons in metals are virtually identical for charged particles whose energy exceeds ≈ 80 eV because the exclusion principle is not a serious limitation on phase space. Aers and Pendry²⁴ used the random-phase approximation (RPA) but warn that it may not be valid for positrons due to enhanced correlation effects. The enhanced damping for positron scattering relative to electrons may be due to the presence of another particle loss channel such as annihilation and elastic positronium formation. Such elastic loss mechanisms are not available for electron scattering. As was recently demonstrated, the incident positron beam approaches the surface at glancing angles, "elastic" positronium formation increases significantly.²⁵

Inelastic processes for positrons inside the solid, however, may be strongly affected by the image cloud of electrons and has not been correctly treated in the theory. For instance, the energy-loss rates for the positive pion and Σ hyperon particles^{26,27} are faster than their associated antiparticles. The disparity has been attributed to the polarization of the target by the incident particle. Qualitatively, the positive projectile attracts electrons inside the solid and thereby increases the collision frequency with the electrons and energy-loss rates. Conversely, negative particles repel electrons and reduce the collision frequency and loss rates. These processes have been examined quantitatively²⁸ and the results should hold for positrons and electrons. The calculation extends the perturbation treatment to the second Born approximation and the stopping power is found to scale as Z^3 where Z is the charge of the incident particle. The experimental results confirm the predictions for more energetic particles than the ones used in the present study. Nevertheless, the calculation of Ashley, Ritchie, and Brandt²⁸ [see Fig. 3 and Eq. (15) in reference 28], indicates that the positron stopping power at 100 eV is enhanced by 50% relative to electrons inside a copper crystal. If the imaginary part of the inner potential β is proportional to the stopping power, then the increase in stopping power of positrons relative to electrons will be reflected in the value of β ($\beta = 6$ eV for positrons and 4 eV for electrons).

Increased damping, however, tends to also broaden the peaks and smear out features. The r -factor analysis requires a large number of maxima and minima for a valid comparison of the spectra from experiment and theory. Therefore, for this particular sample, LEPD I - V curves, relative to LEED, seem to offer a less stringent test of experiment and theory. Weiss *et al.*⁵ suggested that the larger damping of positron waves with respect to electrons would help decrease the computation time due to the reduction of the number of layers in the calculation. Such

an improvement in the calculation was not valid in the present study. A fixed number of layers was used for both positrons and electrons with the relative contribution from each layer determined by the attenuation.

Surface structure determination involves matching the positions and relative heights of the maxima and minima in the predicted and experimental I - V curves rather than achieving agreement in the absolute values. In fact, the absolute values for the experimental spectra are substantially less than the predicted values. Such a difference has been attributed to incoherent elastic scattering of electrons by the steps on the surface. This type of scattering diminishes the intensity of the diffraction spectra and is one source of the diffuse background. Further, the calculation does not deal with the presence of steps although diffuse scattering from a clean crystal has been found to have a very weak dependence on incident beam energy. As suggested by an earlier discussion of the detector, absolute normalization of the I - V curves to the incident beam rate is feasible in the present experimental configuration. The present results suggest that diffuse scattering by steps on the surface for polar angles $\theta \approx 50^\circ$ is similar for positrons and electrons. That is, the ratio between the experimental to the predicted peaks in the I - V curves ranges from 0.2 to 0.5 for both electrons and positrons depending on the particular spectra. The agreement between theory and experiment for the LEPD spectra further suggests that diffuse scattering does not strongly affect the shape of the I - V curves.

For future structural analysis, normal incidence of the positron beam would offer two advantages. As previously mentioned, normal incidence simplifies the angular determination and reduces uncertainty in these important variables. As will be shown in a future publication, positrons tend to enter the solid only superficially when colliding with surfaces at polar angles that are far from normal incidence and therefore the LEPD I - V curve features are broadened under such conditions. Positron diffraction could involve a penetration of the surface intermediate between that of electron diffraction and atom diffraction. Whereas electrons are attracted to the surface by the surface dipole, positrons are repelled from the surface. The static work function for positrons is near zero because the repulsive Coulomb potential acting upon the positron entering the surface is largely balanced by an attractive correlation energy. Positrons with kinetic energies in the range of diffraction experiments should feel less correlation attraction because the electrons cannot follow its rapid motion and hence cannot form a correlation cloud around it. Thus at high energies a positron should be repelled from any surface. This effect should be particularly important at high incidence angles.²⁹ Furthermore, there can be trapping effects in the surface region which cause inelastic scattering and hence a damping of the coherent positron wave, giving a further reduction in penetration.

The surface Cu(100) is well studied and has generally accepted structural and nonstructural parameters. This surface was therefore used as a standard to demonstrate that LEED and LEPD can be employed as a surface probe. Structural analysis of the LEPD I - V spectra sug-

gest that elimination of exchange and retention of the correlation energies yields the best agreement between experiment and calculation. For some crystals such as W(100) (Ref. 2) and Mo(100) (Ref. 3), the approximations of the exchange and correlation terms in the model strongly affect the structural analysis and the values for the fitted LEED derived parameters. LEPD is expected to provide valuable complementary information for the structural analysis of Mo and W due to the absence of exchange and the small correlation energy for positrons inside the refractory metals.

The relative scattering strength for positrons and electrons depends on the scatterer and the incident energy. This fact may be used to elucidate the unresolved structure of ordered overlayers on substrates. We have collected but have not analyzed preliminary LEPD and LEED I - V curves for oxygen on Cu(100) in the $(\sqrt{2} \times \sqrt{2})R 45^\circ$ superstructure. Analysis by a variety of surface probes³⁰ presently disagree among each other on the values for the structural parameters of this particular adsorbate-

substrate system. The preliminary data suggest that positrons like electrons, weakly scatter from the oxygen adsorbate as reflected in the small changes of the 00 and $0\bar{1}$ I - V curve spectral for clean and oxygenated copper and the low intensity of the positron I - V curves for the half-order beams $\frac{1}{2} \frac{1}{2}$ and $\frac{1}{2} \frac{1}{2}$. Further, the LEPD I - V curves from the half-order spots have broad maxima and minima. Other adsorbates may scatter positrons more strongly and LEPD may be a useful tool for studying such systems.

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