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H. L. Davis and J. R. Noonan

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H. L. Davis and J. R. Noonan

Solid State Division, Oak Ridge National Laboratory

Oak Ridge, Tennessee 37830 USA

Abstract

An equivalent beam averaging (EBA) procedure is described which has proved to be very useful to enhance I-V profile data collected for LEED analyses. Specific analyses are documented where application of EBA has led to improved agreement between calculated and experimental I-V profiles. The procedure also has been substantiated by examination of representative I-V profiles which were calculated to correspond to the incident beam slightly misaligned from, and exactly aligned with, the surface normal. It then has been inferred from such substanstiation that use of EBA in a LEED analysis reduces the effects of systematic experimental errors caused by minor misalignment of the incident beam, beam divergence, and certain surface morphologies.

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1. Introduction

To determine a surface's atomic structure by low-energy electron diffraction (LEED) it is necessary to use a trial and error process. In this process experimental Intensity vs. Energy (I-V) profiles for elastically diffracted electron beams are compared with profiles calculated for various geometric models. The surface structure then obtained from a LEED analysis is that assumed structural model which leads to the "best" agreement between calculated and experimental I-V profiles. Since the best agreement is never "perfect," questions can arise concerning the accuracy of an atomic structure determined by a LEED analysis. Also, even if the basic structural model is not questioned, uncertainties will still exist concerning the sensitivity of the analysis to determine, e.g., interatomic bond lengths. So a LEED analysis always is limited by those details which degrade the agreement between experimental and calculated profiles. Thus, it is important that some efforts be made to devise procedures (experimental and/or theoretical model improvement) which can reduce the disagreement between the profiles.

While performing some recent LEED analyses [1-3], we observed that the agreement between calculated and experimental I-V profiles could be improved significantly by the application of a relatively simple procedure. This procedure requires that I-V profile data be collected with the incident electron beam aligned as closely as possible at normal incidence, and then an equivalent beam averaging

(EBA) be performed over sets of as-measured profiles. EBA is explained in more detail below. Some new results then are presented from dynamic LEED calculations for the (100), (111), and (110) surfaces of copper. These calculational results are interpreted to demonstrate that EBA does serve to eliminate some effects of errors that are present in the individual as-measured profiles. In fact, perhaps surprisingly, the calculational results are used to illustrate that EBA serves to eliminate more than the effects of random errors.

2. Description of EBA

Even though a valid LEED analysis would require a data base consisting of several inequivalent I-V profiles, the EBA procedure can be described adequately using, as an example, data [1] for the {11} set of profiles from the Cu(100) surface. These data are illustrated by Fig. 1, where the four as-measured {11} profiles are represented by the four top curves. These {11} as-measured profiles typify the best obtainable from our Cu(100) sample when using our present experimental system [4], and data for all four were collected under as identical experimental conditions as we could control (e.g., after the same surface cleaning and annealing, and the same alignment setting). Since Cu(100) has four-fold symmetry, the four as-measured {11} profiles obtained in a perfect, hypothetical, normal incidence, LEED experiment would be identical by symmetry. Although the as-measured profiles of Fig. 1 are quite similar to each other, some differences do exist

which we believe are due to minor detector misalignment, electron beam divergence, surface topography, residual electromagnetic fields, etc.

The differences between the as-measured profiles can be specified quantitatively using an R-factor as a metric to measure any disagreement. For example, when the Zanazzi and Jona [5] single-beam R-factor, R_{ZJ}, was used to compare various pairs of the as-measured {11} profiles of Fig. 1 with each other, values in the range of 0.04 to 0.10 resulted. Although such values for R_{ZJ} are considered small, it is interesting that they were all larger than the value of 0.027, which resulted when the average of the as-measured profiles (bottom curve of Fig. 1) was compared with the calculated (11) profile from the best case result in our recent [3] analysis of Cu(100).

The final results of a LEED analysis could be influenced by the differences that exist between as-measured profiles that should be symmetrically identical; e.g., the structural conclusion for Cu(100) does have some variation depending upon which of the four top profiles of Fig. 1 is used as data for the analysis [3]. So as an initially pragmatic attempt to reduce the influence of the differences between as-measured profiles, we decided to average profiles that should be symmetrically equivalent. When such EBA was employed in LEED analyses of Cu(100) and Ag(110) [1-3], it was then found that much better agreement was achieved between calculated and averaged experimental profiles than between calculated and as-measured experimental profiles. Thus, the empirical observation emerged that EBA, in some manner, appeared to provide a data enhancement technique for LEED analysis.

In brief summary, the EBA procedure we recommend is to first collect with the incident beam aligned as closely as possible to normal incidence, for each individual {ij} set, all the profiles that should be symmetrically equivalent. A mean profile is then obtained for each {ij} set, and it is the mean profiles that form the data base for the resulting LEED analysis. But as is further discussed below, it is important to realize that not all the differences between profiles that should be symmetrically identical are generated by random errors. Some of the differences have systematic origins. Thus, it is essential that all the profiles in a given {ij} set be used to determine the mean profile, and attempts also should be made to collect the data for all profiles of a given {ij} set under experimental conditions as nearly identical as possible of cleanness, alignment, etc.

3. Calculational Substantiation of EBA

Since EBA has been shown to produce better agreement between calculated and (averaged) experimental I-V profiles, its application in LEED analyses is defensible on a strictly empirical basis. However, it is also possible to justify the application of EBA by some purely calculational arguments. In fact, calculational results now have been obtained whose interpretation directly leads to the conclusion that the improvement gained with EBA is due to the averaging process reducing the effects of some systematic errors. This conclusion can be demonstrated from inspecting the results of performing EBA on I-V profiles purposely calculated for cases where the incident beam is

assumed to be slightly misaligned from normal incidence. To provide examples, illustrative results calculated for the (100), (111), and (110) surfaces of copper are discussed below. These results serve as some calculational substantiation of the use of EBA when performing LEED analyses for surfaces with four-, three-, and two-fold symmetry.

The calculational results represented by Figs. 2-4 were obtained using computer codes based on the layer KKR formalism [6] and RFS perturbation theory [7]. Detailed tests were performed to assure that the calculated results were numerically converged. A total of 8 phase shifts were used, and they were obtained from a truncated free atom potential based on full Slater exchange. For the (100), (111), and (110) surfaces 49, 43, and 47 beams, respectively, were used in the calculations. For all three surfaces, the optical potential was chosen to equal (10+4i) eV. The calculations were performed to correspond to the surfaces being at room temperature, and a Debye temperature of 330 K was employed. For the Cu(100) calculations, the relative relaxations of the first and second interlayer spacings from the bulk value were set at, respectively, -1.10% and +1.70% [3]. For Cu(111), the surface was assumed to be exactly as the truncated bulk (no layer relaxation). Relaxations of -9.0% and +2.0%, respectively, were used for the first and second interlayer spacings in the calculations for Cu(110) [8].

In Figs. 2-4 each of the profiles labeled "average" is the mean of all the profiles above it in the subplot, while the one labeled "normal" was calculated to correspond to the case where the incident beam would be exactly at normal incidence. The profiles labeled with

(ij) beam notation were calculated for the incident beam having an orientation defined by the θ and ϕ angles denoted in the subplots. The (ij) beam indexing is as defined by, e.g., Fig. 5 of Demuth and Rhodin [9]. The angle θ denotes the orientation of the incident beam away from the surface normal, while the incident beam's azimuth is denoted by ϕ. For all three surfaces, tne $\phi = 0°$ azimuth contains the (10) beam. Associated with all but the bottom profile of each subplot of Figs. 2-4 are numbers in the range of 0.010 to 0.160. A given one of these numbers is the single-beam Zanazzi-Jona R_{ZJ} value that resulted when the associated profile was compared with the bottom profile of the subplot (i.e., the one calculated for exactly normal incidence).

First, consider the left subplot of Fig. 2 and note that the differences between the four top profiles are somewhat of the same order as the differences between the as-measured profiles of Fig. 1. Also, note that the R_{ZJ} value for the "average" profile is very small, and the visual agreement between the bottom two profiles of the subplot is much better than between the bottom profile and any of the top four. Thus, the EBA processing of the top four profiles produces an "average" profile in which the effects of systematic errors, caused by a 1° misalignment of the incident beam, have been reduced. It may be noted from the right subplot of Fig. 2 that the EBA processing also significantly reduces the effects of errors caused by a 1.5° misalignment. This reduction has occurred even though the differences between the four top profiles of the right subplot, and between each of the top four and the bottom profile, are much larger than those of the left subplot.

Some results for Cu(111) are illustrated by Fig. 3. Since this surface has three-fold symmetry, the {ij} set for each subplot of Fig. 3 contains only three profiles. Some significant differences are easily seen between the top three profiles of each subplot of Fig. 3, and between the subplot's "normal" profile and specific ones of the top three. However, again the results serve to demonstrate that EBA does reduce the effects of errors caused by minor misalignment of the incident beam. The same type of demonstration is made by the results of Fig. 4, which illustrates some of our results for Cu(110). The {10} set of profiles for Cu(110) is two-fold degenerate, while the {21} set is four-fold degenerate.

4. Conclusions

Besides the calculational results represented by Figs. 2-4, other {ij} sets of profiles, and other cases of slight misalignment from the surface normal, have been investigated for the (100), (111), and (110) surfaces of copper. Values of θ from 0.5° to 2.0°, and various ϕ values, were considered for all three surfaces. Five, six, and eight {ij} sets of beams were considered, respectively, for the (100), (111), and (110) copper surfaces. Also, eight {ij} sets have been considered for Ag(110) for values of θ between 0.5° and 2.0° [10]. Without exception, each set of calculations for a given surface, for specific θ amd ϕ values, and a given {ij} set of beams, provides the same qualitative conclusion as that inferred from the illustrated results of Figs. 2-4. That is, EBA processing of all the profiles in

a calculated {ij} set serves to reduce significantly the effects of errors caused by minor misalignment of the incident beam. Thus, it directly is inferred that using EBA to process as-measured experimental profiles in LEED analyses can reduce significantly the effects of those systematic errors caused by beam misalignment, beam divergence, and certain surface morphologies.

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FIGURE CAPTIONS

Fig. 1. As-measured {11} I-V profiles from Cu(100) and their average. These data were collected with the sample at room temperature.

Fig. 2. Calculated I-V profiles for Cu(100). See the text for a description of how these results substantiate EBA.

Fig. 3. Calculated I-V profiles for Cu(111).

Fig. 4. Calculated I-V profiles for Cu(110).

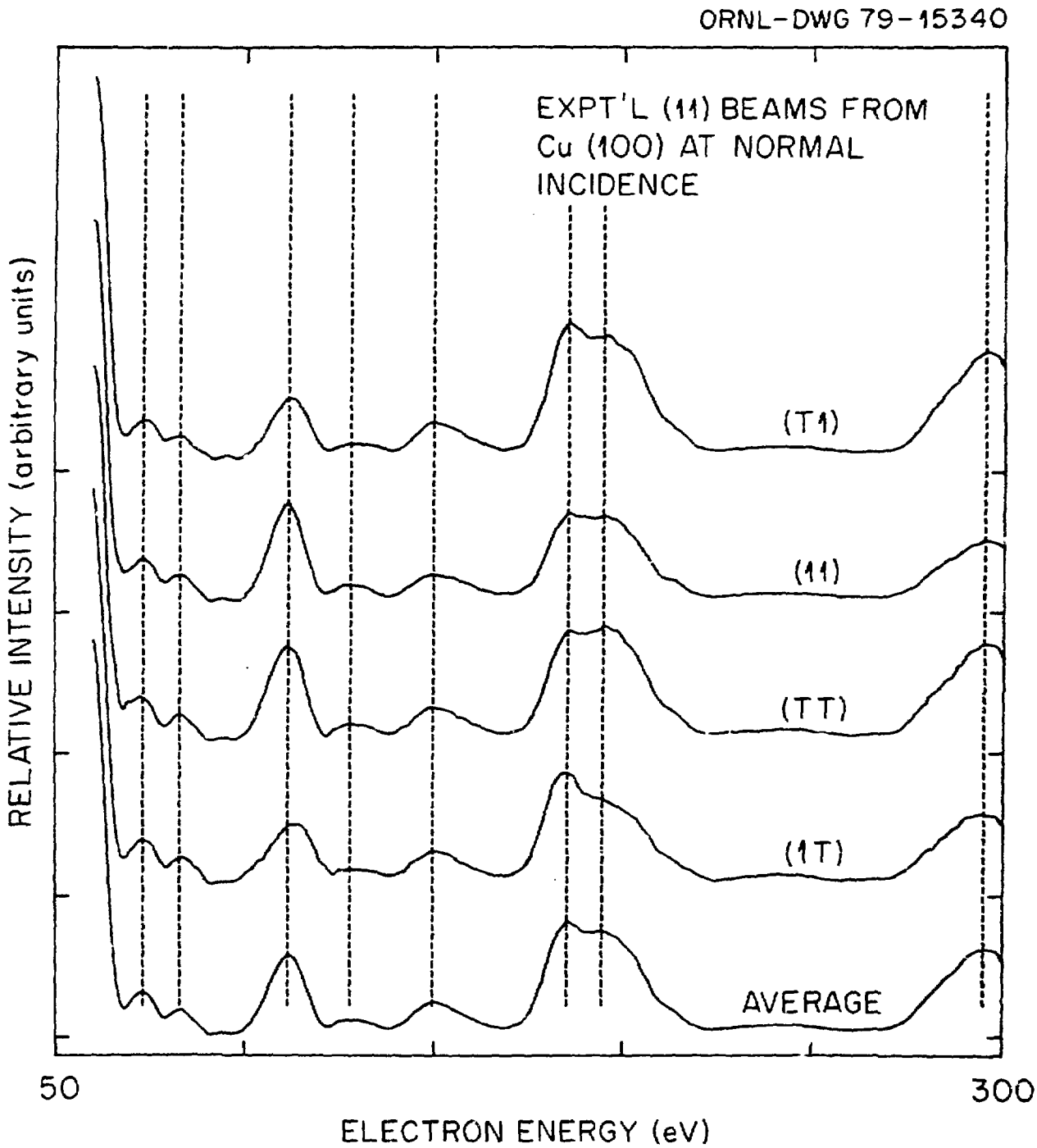
ORNL-DWG 79-15340
EXPT'L (11) BEAMS FROM
Cu (100) AT NORMAL
INCIDENCE
(T1)
(11)
(TT)
(1T)
AVERAGE
RELATIVE INTENSITY (arbitrary units)
50
300
ELECTRON ENERGY (eV)

Fig. 1

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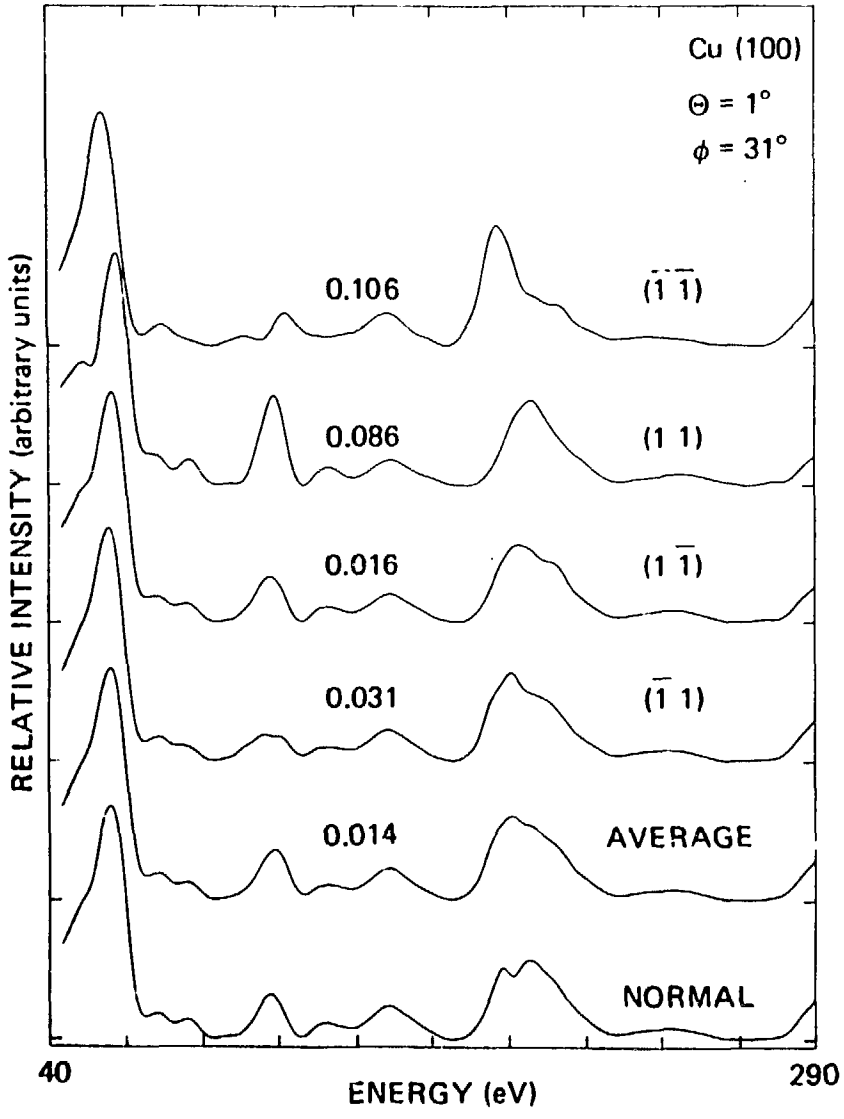

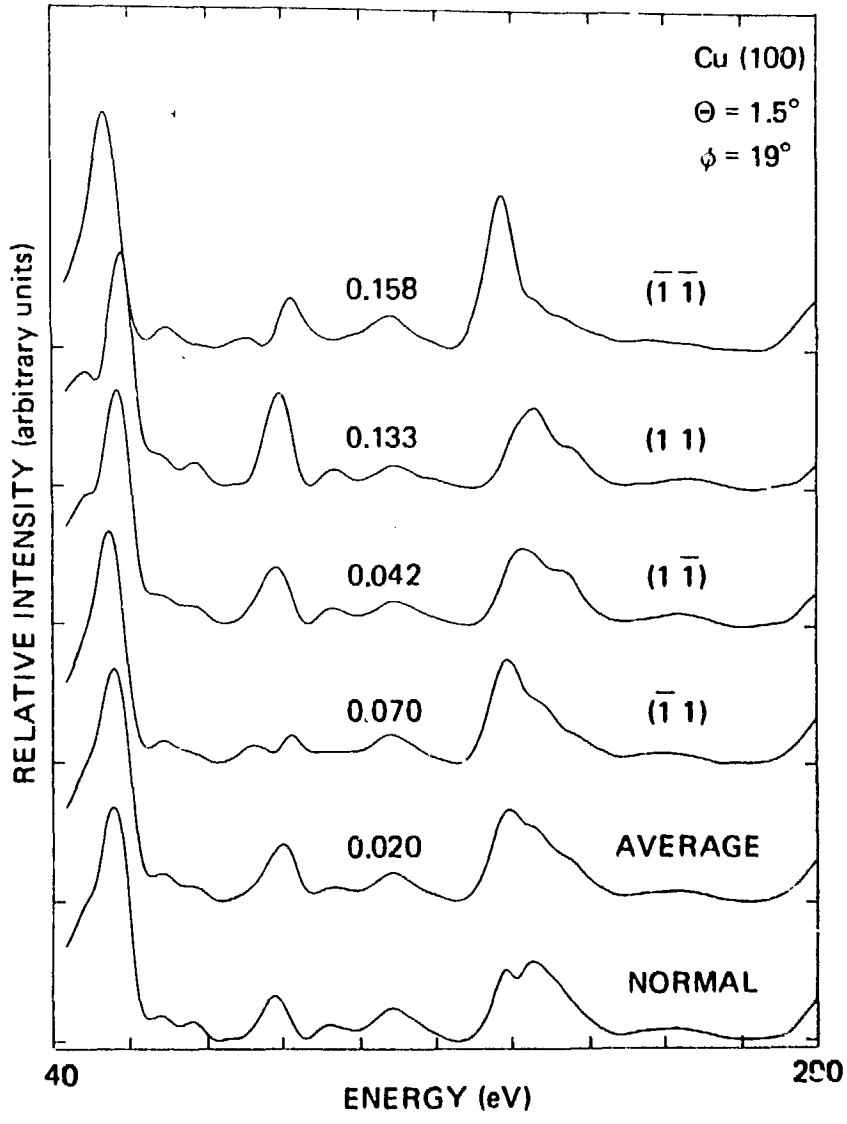

Fig. 2

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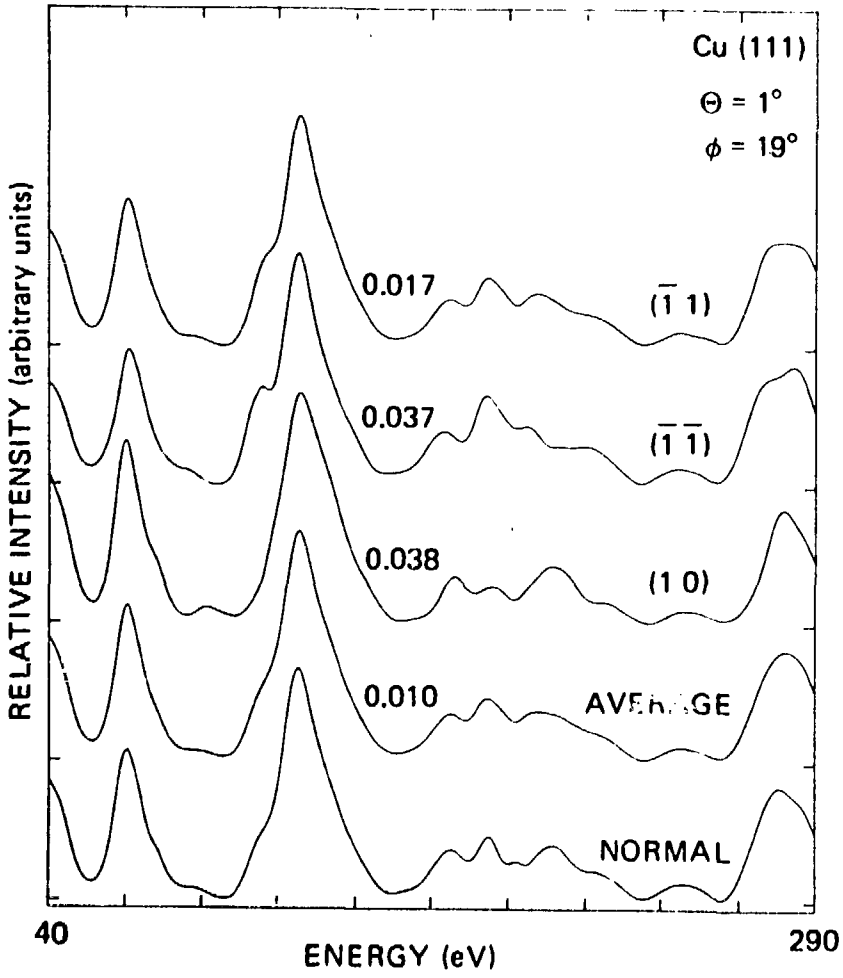

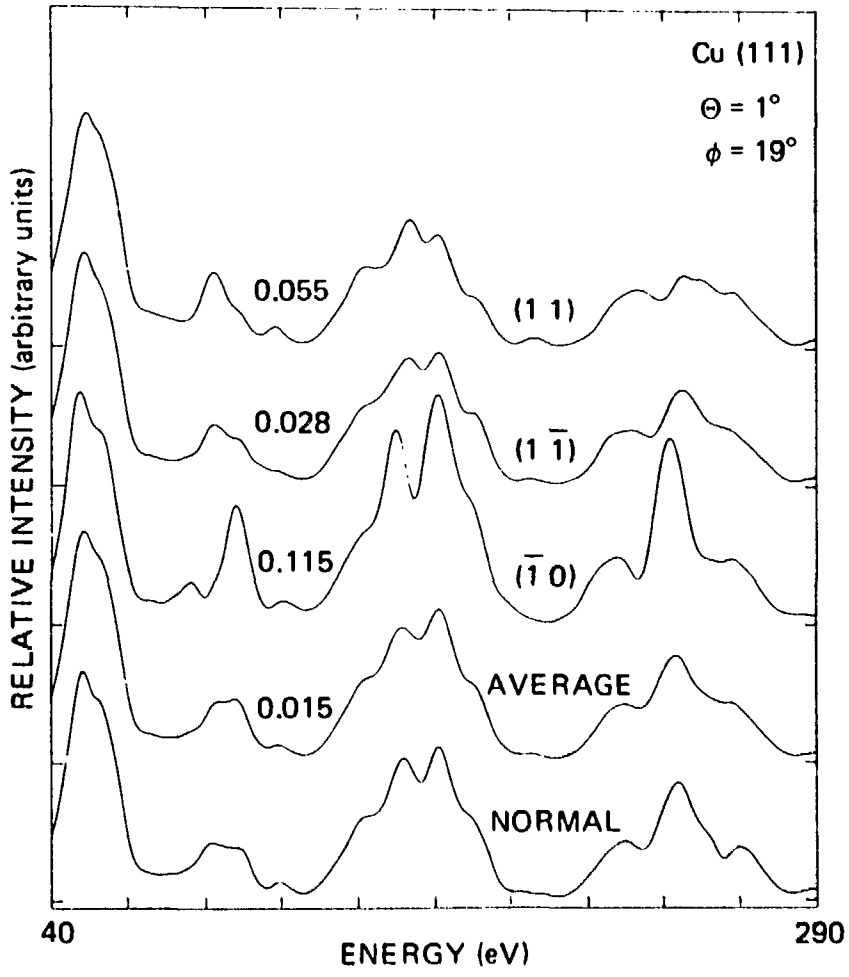

Fig. 3

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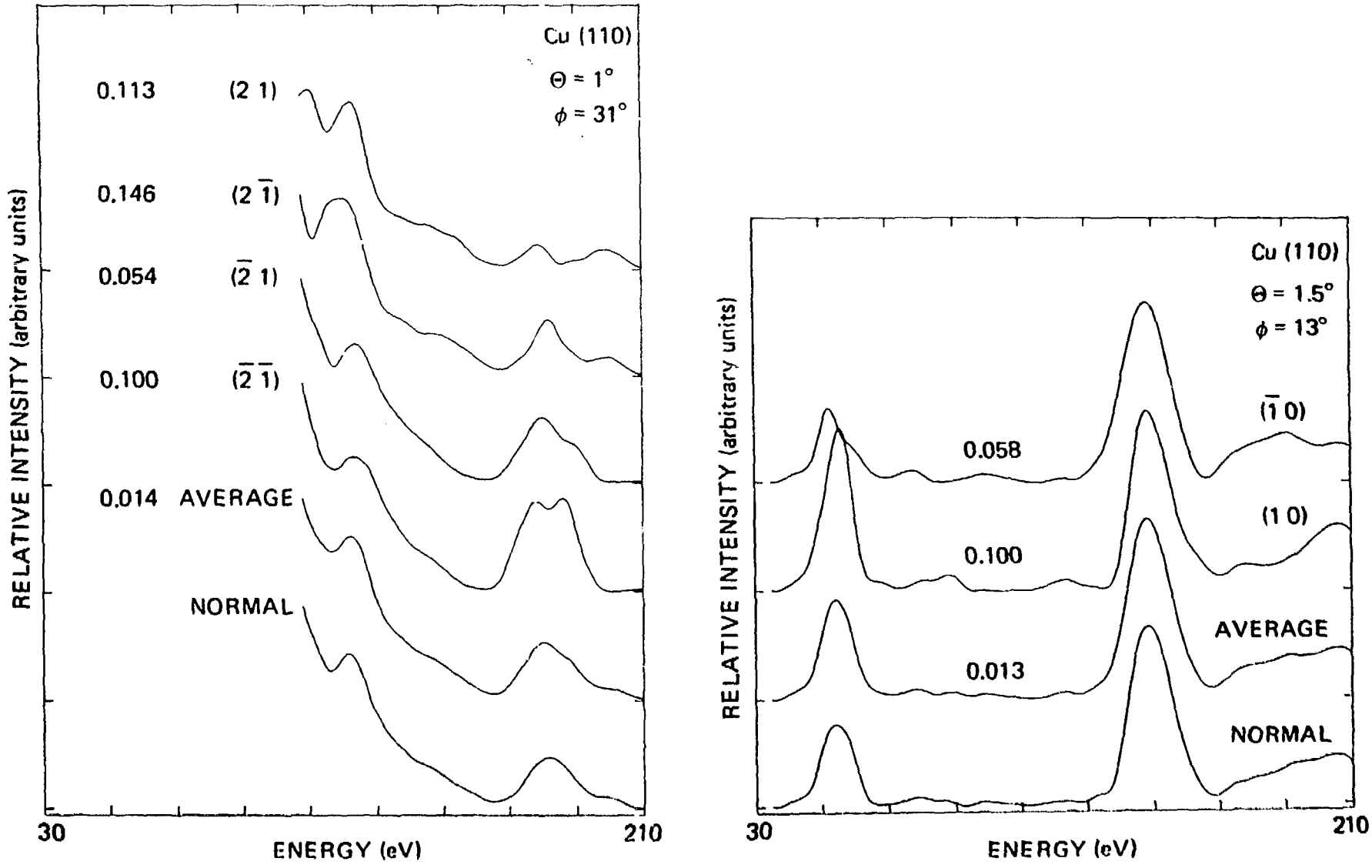

Fig. 4