Auger Electron Angular Distributions from Surfaces: Forward Focusing or Silhouettes?

The recent article by Douglas G. Frank et al. (1) contains a number of conceptual errors that undermine both the data interpretation and the conclusions. The authors base their analysis on an erroneous set of notions about electron atom scattering. At the center of their misunderstanding is the statement that Auger emission events are uncorrelated and therefore cannot undergo the "formation of plane waves required for efficient diffraction." They then go on to imply that photoemission events are correlated and that the resulting photoelectrons can thereby undergo coherent diffraction. In point of fact, both Auger emission and photoemission events are uncorrelated. Also, correlation between Auger or photoemission events is not required to realize diffraction. Diffraction is nothing more than elastic scattering and interference, and all that is required for Auger or photoelectron diffraction to be detectable in an angle-resolved measurement is that the emitter be situated in a single crystal. In such a situation, the observed angular distributions show considerable intensity modulation as a result of the interference of the unscattered wave portion and all elastically scattered wave portions at the detector point.

Frank et al. then go on to claim that the modulation is caused entirely by "shadowing" (inelastic scattering). Yet, elastic scattering cross sections show considerable angular dependence and are largely peaked in the forward direction for all but the lowest kinetic energies. In contrast, there is no convincing evidence that inelastic scattering of low to medium kinetic energy electrons at single-crystal surfaces shows any anisotropy. The primary loss mechanism is plasma excitation, which is largely delocalized. A much weaker loss mechanism is excitation of bound core states by dipole scattering, which might be expected to show some angular dependence. In making their assertions, Frank et al. tacitly ignore 10 years of successful application of elastic scattering theory to the interpretation of angle-resolved Auger and photoelectron spectroscopic data. In rationalizing their results, they also ignore the well-established fact that Auger electrons and photoelectrons of the same kinetic energy from the same specimen exhibit nearly identical angular distributions, all of which are very well predicted by elastic scattering theory (2).

So why do the data of Frank et al. show minima along interatomic directions in Pt(111)? One possibility is that their specimens were not properly oriented about the surface normal during the measurements (3). If their crystal was rotated 60° about the surface normal relative to where they thought it was, the low-energy electron diffraction pattern would not change, but the Auger intensity pattern would be inverted. If this error was made, what was interpreted to be electron intensity poking through the spaces between surface atoms would actually be forward-scattering-inelastic maxima along interatomic vectors. The latter interpretation is certainly much more consistent with basic principles of quantum-mechanical scattering than the proposition they forwarded.

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3. S. Y. Tong, personal communication.

23 January 1990; accepted 11 April 1990

The physics underlying the angular distribution of Auger electrons is well understood (1), having been the subject of study for two decades. However, the recent article by Frank et al. (2) dismisses as mistaken virtually the whole of this body of work. We believe that the work of Frank et al. is mistaken, that the well-established models are the correct ones, and that the origin of this dispute is a very limited, unrepresentative data set that Frank et al. have interpreted with models that contain a mixture of gross oversimplification and error.

The crux of the interpretation by Frank et al. is that atoms cast shadows so that Auger intensities are weak along interatomic directions. As a general proposition this is false. For Auger electrons with a kinetic energy below ~100 eV, the angular distribution of the intensity is observed experimentally to be a strong function of kinetic energy. The directions in which high and low intensities are observed are different for Auger electrons with different kinetic energies (3), and this is incompatible with the model proposed by Frank et al. Moreover, the Auger electrons from different materials with a common crystal structure give different angular distributions (4). These results are due to complicated diffraction phenomena involving multiple elastic scattering of the emitted Auger electron, effects that are highly energy dependent. Therefore the approach to determining surface structure that Frank et al. suggest has no general validity.

At kinetic energies of a few hundred electron volts and above, Auger angular distributions are well known to exhibit enhanced intensities along interatomic directions because of forward scattering (or forward focusing). In the past decade this effect has been developed into a useful tool for surface structural determination (5).

Other errors in this paper are too numerous for a complete list here, but among the most egregious are the claims that only 4% of the scattering events are elastic and that it is the bound electrons (rather than the total atomic potential) that scatter an incident Auger electron.

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29 January 1990; accepted 11 April 1990

Frank et al. (1) claim that Auger electrons emitted from atoms in deeper layers have angular profiles that peak along the spaces between surface atoms. These authors explain their result as surface atoms casting shadows or silhouettes on the emission from deeper layer atoms. In other words, the surface atoms block the transmission of Auger electrons along interatomic directions. They claim that the shadowing effect is generally valid and that this effect presents a direct method for imaging surface atomic structure. In contrast, previously published
models of Auger electron angular distributions have found intensity maxima lying along interatomic directions, precisely the opposite of Frank et al. results (2). Frank et al. claim that this contradiction is due to a lack of complete angular distribution data in the previous studies.

We have taken similar full-scan angular distributions by Auger electron spectroscopy (AES) at 914 eV and by x-ray photoelectron spectroscopy (XPS) at 1401 eV from Cu(111) (3), as well as by AES at 917 eV from Cu(001) (4). These AES and XPS data show a common trend in that the intensity maxima are directed along interatomic axes. Thus, instead of surface atoms blocking the transmission of Auger electrons from deeper layers, the surface atoms actually enhance Auger (or XPS) transmission along interatomic directions. In Fig. 1 we show our AES and XPS data for Cu(111), along with the crystallographic directions, and a map of the arrangement of near-surface atoms in real space. In Fig. 1, the most intense AES

The correct interpretation of the intensity enhancement of AES or XPS emission along interatomic axes observed at high energies is the strong forward scattering of the emitted electrons as they pass near the (attractive) atomic-core potential of a surface atom (5, 6). This phenomenon has been likened to the focusing of an isotropically diverging beam of electrons into directions parallel to the interatomic axis. The underlying physical processes of forward focusing are shown in Fig. 2A. Forward focusing not only correctly explains the observed azimuthal directions of the intensity maxima but also quantitatively explains the polar angles at which the intensity maxima point (2-9). For example, the Auger electron (and XPS) angular-distribution data (Fig. 1) have intensity maxima along polar angles $0^\circ$, 19.5°, and 54.7° along the [112] azimuth and at $35.3^\circ$ along the [112] azimuth, which is the most intense peak. Each of these intensity peaks corresponds exactly with a high-density interatomic axis in the near-surface region of the Cu(111) surface (Fig. 2B). The forward-focusing peaks in the AES and XPS data are well reproduced by multiple-scattering theory (3, 6, 7). (see Fig. 1). The physics of forward focusing is now well understood and these results have been corroborated by other workers. A number of extensive reviews are now available (2, 9).

How, then, do the data of Frank et al. fit into this picture? Their Pt(111) Auger distributions were taken at a very low kinetic energy. Frank et al. chose this energy to maximize the intensity of the signal. As we have pointed out (10), forward focusing is valid only for kinetic energies of electrons above a few hundred electron volts. In an exact quantum mechanical treatment of electron scattering from Cu atoms, the forward-focusing enhancement becomes strong and independent of energy only if the kinetic energy is above 300 eV (10) [see figure 5, a through d, of (10) for the dependence of the forward-scattering cone on kinetic energy]. The Pt(111) data by Frank et al. may be

![Fig. 1.](image)

![Fig. 2.](image)
explained by the fact that in the low-energy region the Auger angular distribution is a rapid function of kinetic energy. Thus the result they obtained was specific to a particular kinetic energy and has no general validity. The agreement they obtained with a model with "shadowing" was accidental and was directly opposite to the physical concept and trend at higher energies. Another possible explanation of the results of Frank et al. is that they made an error of 60° as they superimposed their data on the real-space crystallographic directions. The crystallographic directions indicated in our work have been independently determined by x-ray scattering and by analysis of low-energy electron diffraction intensity voltage curves.

Angular distributions for AES and XPS provide a map of intensity enhancements along high-density interatomic directions, provided that the kinetic energy of the emitted electrons is high enough (for example, above a few hundred electron volts). Enhanced forward scattering (or forward focusing) is the correct physical explanation for the general trend. Auger angular distributions at very low energies are energy-dependent and hence they do not have a single fixed relation to the surface structure. Explanation of the general trend in terms of shadowing is wrong and is not supported by data at high energies.

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It is an important principle of scientific development that new theories should account for both new and old experimental data. It is therefore disturbing to see the recent article by Frank et al. (1) concerning the angular dependence of Auger electron emission from solids that claims to have developed a new theory while dismissing older theories as wrong and totally failing to consider a wealth of old experimental data that support the "old" theories. Frank et al. (1) discuss the influence of atoms surrounding an Auger electron emitter (at kinetic energies of 65 and 355 eV) purely in terms of local "shadowing." Such a treatment totally neglects the quantum mechanical wave nature of the electrons in this energy range; indeed, this is precisely the energy range in which Davisson and Germer (2) first demonstrated the wave nature of electrons through scattering by atoms in the surface of a solid (a closely related phenomenon) for which they received the Nobel Prize.

Despite this fundamental flaw in the starting point, it is striking that the "theory" of Frank et al. appears to fit the data rather well; this success, I believe, can be understood in terms of the proper quantum mechanical description, which is far more widely applicable. In particular, two features characterize electron scattering by atoms in this energy range. First, there is invariably a peak in the forward-scattering amplitude; but second, the scattering factor is complex, so there is a phase shift between the directly transmitted and forward-scattered component. If this phase shift is close to $\pi$, the interference between these two components is destructive and a reduced (shadowed) forward-scattering intensity is seen. This effect is most common at low energies and is probably the main qualitative effect in the data of Frank et al. (although the data can only be modeled reliably by adding in many scattering events). However, if the phase shift is small compared with $\pi$, the interference is constructive and enhanced forward scattering ("focused") intensity is seen. This effect is the usual state of affairs at high energies (above $\sim 500$ eV). Perhaps the nicest example of this effect (the opposite of that seen by Frank et al.) is in photoemission, rather than Auger electron emission, from a diatomic molecule such as CO (3). It is also seen in studies of epitaxial layer growth (4). For chains of atoms (as in the Pt case of Frank et al.), the situation is rather more complex because multiple forward scattering along the chain can lead to either enhanced or attenuated emission emerging from the chain.

Finally, I should remark that this phenomenon of coherent interference of elastically scattered electron wavefield components can also occur in backscattering. The effect is weak at high energies (for example, the 518-eV iodine Auger emission in the work of Frank et al.), so little angular dependence is seen. However, at low energies strong Auger (and photoemission) angular features are seen from this effect from atoms that lie above all of the scattering atoms (5) and are therefore totally inexplicable within the framework of the Frank et al. theory.

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21 February 1990, accepted 11 April 1990

Response: In our recent article (1) we reported that angular distributions of Auger electrons emitted from single-crystal surfaces and monolayers contain the silhouettes of surface atoms backlit by emission from atoms deeper in the solid. Simulations based upon atomic point emitters and spherical atomic scatterers of Auger electrons were in close agreement with the experimental results. Angular distribution Auger microscopy (ADAM) is a useful technique for direct imaging of interfacial structure as well as for investigating the interaction of electrons with matter. Applicability of ADAM was illustrated by images obtained for platinum (111) (Pt[111]) and for monoatomic layers of silver and iodine on Pt[111]; several other samples have also been imaged.

Briefly, our conclusions are that atoms behave as isotropic point emitters and spherical scatterers of Auger electrons and that inelastic scattering predominates over elastic scattering at low kinetic energies, leading to relatively simple, surface-sensitive images. However, certain experimental criteria must be met in order to obtain useful ADAM images, including (i) the direction of the incident beam in relation to the sample must be held constant to eliminate one of the two complicated geometric effects in the experiment; (ii) modulation of pass energy and synchronous detection in order to distinguish Auger electrons from the much more numerous background electrons; (iii) angular accuracy and precision better than $\pm 1^\circ$ to locate the many sharp features of typical
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Science 248 (4959), 1129-1131.
DOI: 10.1126/science.248.4959.1129-b