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 specimen was 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–induced 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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REFERENCES

1. D. G. Frank *et al.*, *Science* **247**, 182 (1990).
2. P. J. Orders, R. E. Connelly, N. F. T. Hall, C. S. Fadley, *Phys. Rev. B* **24**, 6163 (1981); M. Owari, M. Kudo, Y. Nihei, *J. Electron Spectros. Relat. Phenom.* **22**, 131 (1981); S. A. Chambers and T. J. Irwin, *Phys. Rev. B* **38**, 7858 (1988).
3. S. Y. Tong, personal communication.

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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 $\sim$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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REFERENCES

1. M.-L. Xu, J. J. Barton, M. A. Van Hove, *Phys. Rev. B* **39**, 8275 (1989); M.-L. Xu and M. A. Van Hove, *Surf. Sci.* **207**, 215 (1989).
2. D. G. Frank, N. Batina, T. Golden, F. Lu, A. T. Hubbard, *Science* **247**, 182 (1990).
3. T. Matsudaira and M. Onchi, *Surf. Sci.* **74**, 684 (1978); L. McDonnell, D. P. Woodruff, B. W. Holland, *ibid.* **51**, 249 (1975); D. M. Zehner, J. R. Noonan, L. H. Jenkins, *Phys. Lett.* **62A**, 267 (1977).
4. G. Allié, E. Blanc, D. Dufayard, *Surf. Sci.* **57**, 293 (1976); D. Aberdam, R. Baudoing, E. Blanc, C. Gaubert, *ibid.* **71**, 279 (1978); J. D. Place and M. Prutton, *ibid.* **82**, 315 (1979).
5. For a review of this field, see W. F. Egelhoff, Jr., *CRC Crit. Rev. Solid State Mater. Sci.* **16**, 213 (1990).

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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 (or XPS) emissions are pointed along the $[11\bar{2}]$, $[1\bar{2}1]$, and $[\bar{2}11]$ directions, whereas the "shadowing" theory of Frank *et al.* would have these maxima pointing toward the opposite directions, that is, $[\bar{1}\bar{1}2]$, $[\bar{1}2\bar{1}]$, and $[2\bar{1}\bar{1}]$, respectively [see figure 5C in (*1*)].

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 peaks pointing along polar angles $\theta = 0°$, $19.5°$, and $54.7°$ along the $[\bar{1}\bar{1}2]$ azimuth and at $\theta = 35.3°$ along the $[11\bar{2}]$ 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

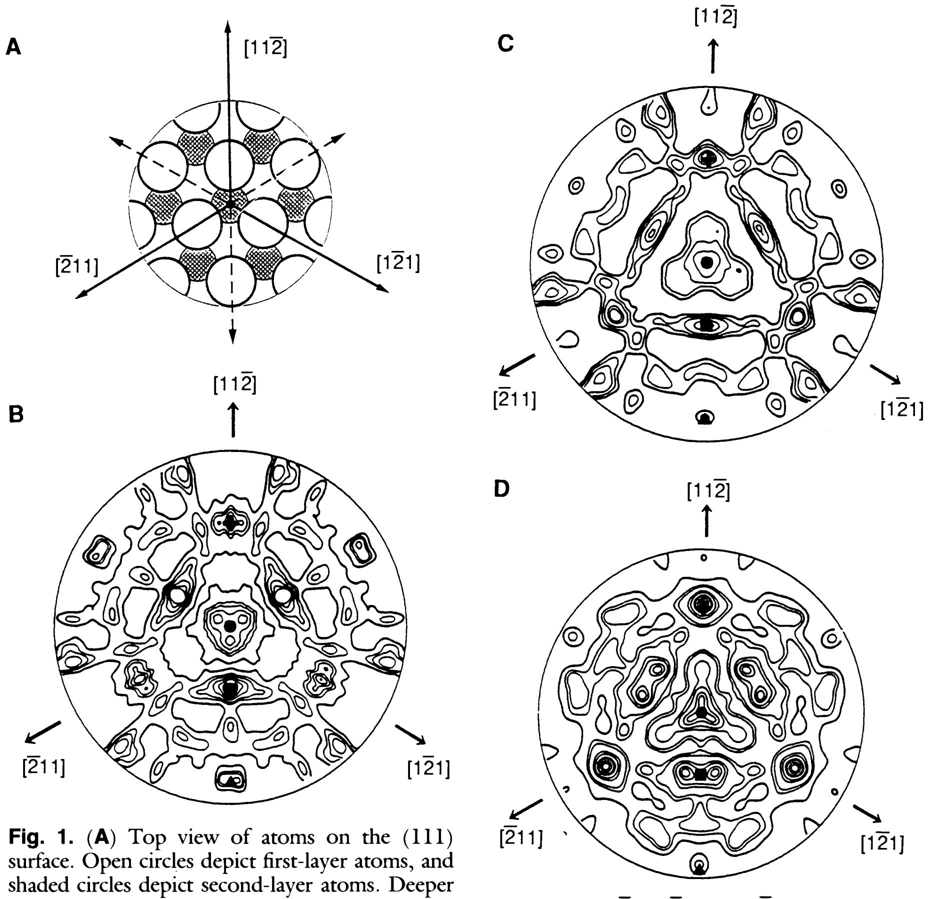

Fig. 1. (**A**) Top view of atoms on the (111) surface. Open circles depict first-layer atoms, and shaded circles depict second-layer atoms. Deeper layers not shown. The most intense emission peaks are along the $[11\bar{2}]$, $[1\bar{2}1]$, and $[\bar{2}11]$ azimuths. (**B**) Contour map of AES angular-distribution data for Cu(111) [kinetic energy (*KE*) = 914 eV]. The major intensity peaks are indicated by the crystallographic directions shown: (+), [110], $\theta = 35.3°$ (most intense peak); (●), [111], $\theta = 0°$; (■), [112], $\theta = 19.5°$; and (▲), [001], $\theta = 54.7°$. The + direction corresponds exactly to the most dense atomic axis in the crystal. Polar-angle range is 70°. (**C**) XPS angular-distribution data for Cu(111) (*KE* = 1401 eV) showing major peaks pointing along exactly the same crystallographic directions as in the AES angular-distribution data. Polar angle range is 70°. (**D**) Calculated contour XPS map (*KE* = 1401 eV) based on multiple scattering theory. The theory reproduced the directions of all major intensity peaks in the data. Polar angle range is 60°.

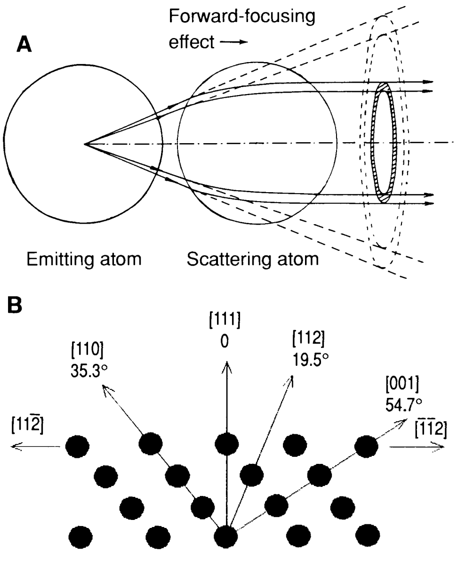

Fig. 2. (**A**). Diagram depicting increase of Auger (or XPS) electrons along directions parallel to the interatomic axis due to forward scattering of electrons by an attractive Coulomb potential of a neighboring atom. Broken radial lines denote directions along which the intensity is decreased from that of an isolated emitter. (**B**) Side view of Cu(111) showing crystallographic directions along which the density of scatterers (atomic cores) are high. These directions have a one-to-one corespondence to the AES or XPS intensity-maxima shown in Fig. 1 as well as to those calculated by multiple-scattering theory.

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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REFERENCES

1. D. G. Frank, N. Batina, T. Golden, F. Lu, A. T. Hubbard, *Science* **247**, 182 (1990).
2. W. F. Egelhoff, Jr., CRC *Crit. Rev. Solid State Mater. Sci.* **16**, 213 (1990).
3. X. D. Wang *et al.*, *Bull. Am. Phys. Soc.* **35** (no. 3), 443 (1990).
4. H. Li and B. P. Tonner, *Phys. Rev. B* **37**, 3939 (1988).
5. H. C. Poon and S. Y. Tong, *ibid.* **30**, 6211 (1984).
6. S. Y. Tong, H. C. Poon, D. R. Snider, *Phys. Rev. B* **32**, 2096 (1985).
7. M. L. Xu and M. A. Van Hove, *Surf. Sci.* **207**, 215 (1989).
8. H. Li and B. P. Tonner, *Phys. Rev. B* **40**, 10241 (1989); S. A. Chambers, S. B. Anderson, J. H. Weaver, *ibid.* **32**, 4872 (1985).
9. C. S. Fadley, in *Synchrotron Radiation Research: Advances in Surface Science*, R. Z. Bachrach, Ed. (Plenum, New York, in press); S. Y. Tong, M. W. Puga, H. C. Poon, M. L. Xu, in *Chemistry and Physics of Solid Surfaces*, R. Vanselow and R. Howe, Eds. (Springer, Berlin, 1986), vol. 6, pp. 509–545.
10. H. C. Poon, D. R. Snider, S. Y. Tong, *Phys. Rev. B* **33**, 2198 (1986).

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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 π, 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 π, 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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REFERENCES

1. D. G. Frank, N. Batina, T. Golden, F. Lu, A. T. Hubbard, *Science* **247**, 182 (1990).
2. C. Davisson and L. H. Germer, *Phys. Rev.* **30**, 705 (1927).
3. L.-G. Petersson, S. Kono, N. F. T. Hall, C. S. Fadley, J. B. Pendry, *Phys. Rev. Lett.* **42**, 1545 (1979).
4. W. F. Egelhoff, Jr., *J. Vac. Sci. Technol. A* **3**, 1511 (1985).
5. D. A. Andrews and D. P. Woodruff, *Surf. Sci.* **141**, 31 (1984); D. P. Woodruff *et al.*, *Phys. Rev. Lett.* **41**, 1130 (1978).

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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 ±1° to locate the many sharp features of typical

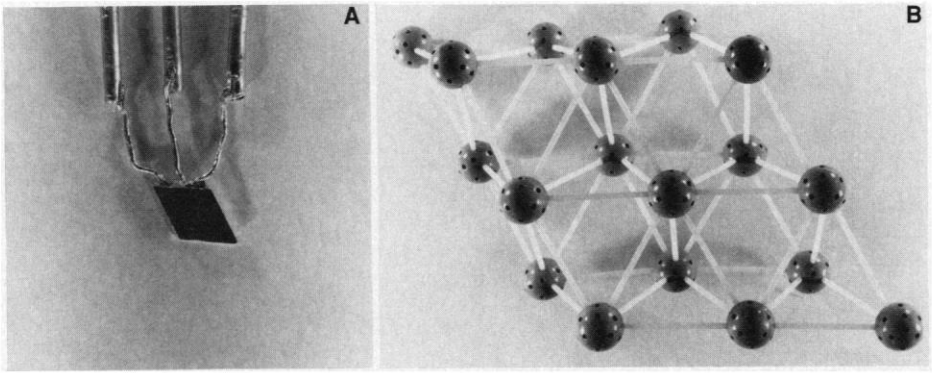

Fig. 1. (**A**) Photograph of the platinum single crystal used for these studies. All six faces were oriented and polished parallel to the [111] or equivalent crystallographic planes. The crystal is shown in the orientation studied. (**B**) Model of Pt[111] shown in the same orientation as the crystal in (A).

distributions; (iv) signal-to-noise ratios greater than 50:1 to preserve subtle features; (v) kinetic energy resolution sufficient to separate competing Auger lines of substrate and overlayer; and (vi) scanning the complete range of angles above the surface so as to define unambiguously the nature of the distribution.

The correspondents object to the ADAM images containing minima where they expected maxima based upon their "forward-focusing" hypothesis. To rationalize the failure of their "forward-focusing" model, they suggest that our Pt[111] sample was misaligned. However, as is standard practice in our laboratory, "the Pt[111] single crystal was oriented and polished such that all six faces were crystallographically equivalent," [references 2 and 34 of (*1*)]. This extra care in preparation of the crystal, which includes Laué photography of each of the six crystal faces, permits its use as an immersed electrode and also makes its orientation unmistakable. A photograph of the crystal used in the experiments is shown in Fig. 1A, and a model having the same orientation as our sample is shown in Fig. 1B. Note that the left and top edges are visible in this orientation and that the silhouettes of atoms located in the top layer are present at the correct locations in the ADAM image [figure 5 of (*1*)]. "Forward focusing," which predicts maxima where minima occur, is not observed.

Another reason that the correspondents are surprised by our data is that they incorrectly assume that inelastic scattering in a crystal is homogeneous. Indeed, it is a dubious assumption that a crystal, composed of a periodic array of atoms, would behave as "jellium" when inelastically scattering electrons. Also, it is a mistake to assume that elastic scattering processes would predominate, given the wealth of electron spectroscopic evidence to the contrary that clearly demonstrates the importance of energy-loss processes. As illustrated by the data in figure 1B of (*1*), elastic scattering typically amounts to a few percent of scattering events. Furthermore, elastic backscattering actually contributes to the silhouettes seen in ADAM images. Substrate Auger signals are attenuated by about 30% due to the presence of a single monoatomic overlayer. Thus it should not be surprising that Auger emission angular distributions would reveal crystalline inhomogeneity when measured with sufficient precision and angle range at constant incident beam direction for well-characterized samples at low kinetic energies.

The correspondents claim that emission of Auger electrons from atop atoms is strongly anisotropic and that the observed isotropic distribution from adsorbed iodine (507 and 518 eV) in figure 6B of (*1*) is an exception because of its high energy. However, we have measured the angular distributions of Auger electrons fron monoatomic layers of silver (355 eV), chlorine (181 eV), and sulfur (152 eV), all of which are isotropic to within the precision of our measurements (about ±2%). Likewise, the Pt[111] Auger distribution at 65 eV is accurately described by equations based upon isotropic emitters. Therefore, it is not obvious what caused the angular variations reported by the correspondents. A possible source of those variations is that the angle of the incident beam relative to the sample was varied in their experiments. Regarding this undesirable complication, it has been reported (*2*) that "a minor variation of angle of incidence, θ_i, for instance of 1° around $\theta_i = 45°$ can lead to a dramatic change in the relative peak heights. Hence one is led to the conclusion that the information contained in a single spectrum or in a small number of spectra cannot possibly suffice for a consistent picture which correlates well defined losses with certain structures in those spectra."

The correspondents express the viewpoint that our results, which contradict their interpretations, are "limited and unrepresentative" and that their explanation works better at higher kinetic energies, where they claim to have found "enhanced intensities along interatomic directions." We presented ADAM images based on Pt Auger emission at 65 eV and silver at 355 eV. Thus the data presented are typical of the energy range used in Auger experiments. Also, as we explained, lower kinetic energies are preferable because they produce simpler, more surface-sensitive images. For example, the principal interatomic axes are located in the three largest atomic silhouettes at $\phi = 35.3°$ with $\theta = 30°$, 150°, and 270° [see figure 5C in (*1*)]. Note that intensity maxima are *not* observed along those directions. There is, however, a faint rhombic formation of higher intensities surrounding that region, the [110] normal. This feature predominates at higher kinetic energies (*3*) and is not precisely along, but near, the internuclear direction normal to [110]. Thus, if angular resolution, angular accuracy, or data density are insufficient, such features can be mistakenly assigned, leading to incorrect interpretations.

The correspondents suggest that we are "totally failing to consider" or are "unaware" of their data and interpretations. Not only are we aware of their work, but we cited a generous sampling in (*1*) (references 1 through 20); and although we agree with the conclusions of references 1 through 4, we pointed out basic experimental and interpretational shortcomings of references 5 through 20:

1) complete angular distributions spanning the full hemisphere were not measured, simulated, or displayed; analyzer angular resolution and data density with respect to angle were insufficient;

2) the direction of the incident beam with respect to the sample surface was not held constant;

3) distributions were measured at higher than optimal kinetic energies, which complicated interpretation;

4) signals were often not differentiated with respect to energy (dN/dE) in order to distinguish Auger electrons from background;

5) data were interpreted in terms of anisotropic Auger electron emission from individual atoms;

6) inelastic scattering of Auger electrons by crystals was assumed to be homogeneous; and

7) elastic scattering and multiple elastic scattering were overemphasized.

It seems that the correspondents' primary objection is that we have not used their "forward-focusing" model to explain our results. As noted in (*1*), the forward-focusing model does not adequately describe our angular distributions of Auger electron emission: (i) intensity maxima are not observed where "forward-focusing" models would predict them to be; (ii) observed maxima correspond instead to gaps or channels between atoms, sometimes located near but seldom directly along the interatomic axes; and (iii) atomic scatterers become smaller and more transparent at higher kinetic energy; our model continues to account for these results. Obviously, electrons exhibit a duality of wave and particle properties, and evidently, the ADAM experiment emphasizes the particle properties. One of the correspondents suggests that perhaps the observed silhouettes are due to an interference effect resulting in multiple scattering such that atomic scatterers produce minima for kinetic energies below about 500 eV but maxima otherwise. However, our experimental results do not support this idea; a monoatomic layer of scatterers (for which multiple scattering is especially improbable) produces distinct silhouettes even at energies near 500 eV.

The correspondents assert that Auger and photoelectron events are uncorrelated. As stated in (*1*), we agree that Auger events are uncorrelated but we leave open the possibility that coherent photoemission might be observable under some circumstances. That is, although the rates of Auger and photoelectron processes "have not been measured in the laboratory" (*4*), estimates place Auger processes "in the 10^{-16}- to 10^{-15}-s range," whereas the photoelectron process is generally considered to be "faster than 10^{-16} s" (*5*).

We believe that the correspondents' objections are based on faulty assumptions and are contradicted by the experimental evidence. We hope that recent developments will rekindle interest in the interaction of electrons with matter, an important area that is not at all "well understood." Prospects for future discoveries and practical applications in the area are excellent, provided that the appropriate experimental criteria are met in future work. Applications to epitaxial deposition, crystallography, alloys and materials, superconductivity, surface characterization, electrochemistry, and a wide variety of other areas are likely.

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REFERENCES AND NOTES

1. D. G. Frank, N. Batina, T. Golden, G. Lu, A. T. Hubbard, *Science* **247**, 182 (1990).
2. G. Meister *et al.*, *Surf. Sci.* **143**, 547 (1984). See also L. H. Harris, *ibid.* **15**, 77 (1969); J. J. Vrakkong and F. Meyer, *ibid.* **35**, 34 (1973); L. H. Tjeng, R. W. Bartstra, G. A. Sawatzky, *ibid.* **211/212**, 187 (1989); F. Peeters, E. R. Puckrin, A. J. Slavin, *J. Vac. Sci. Technol.* **A8**, 797 (1990).
3. D. G. Frank *et al.*, *Langmuir* **5**, 1141 (1989).
4. F. Brown, *Solid State Phys.* **29**, 1 (1974).
5. D. Coulman *et al.*, *J. Chem. Phys.*, in press; see also D. Coulman *et al.*, *Chem. Phys. Lett.* **143**, 371 (1988).

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"I think she runs it flawlessly once in a while just to louse up our statistics."