## BACKSCATTERING COEFFICIENTS FOR LOW ENERGY ELECTRONS

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(Received for publication August 6, 1996 and in revised form December 9, 1996)

#### **Abstract**

We present new experimental results of the backscattering coefficient ( $\eta$ ) for C, Al, Cu, Ag and Au targets bombarded with low energy electrons (0.6-6 keV) at normal angles of incidence. The present data show a clear monotonic increase in the backscattering coefficient with increasing target atomic number for electrons of primary energy less than 1 keV in contrast with previously published data. A fundamental difference between this study and other reported measurements is that the present data set is collected from *in situ* cleaned surfaces under ultra high vacuum conditions ( $10^{-10}$  mbar), while the previously reported studies were carried out under conventional vacuum ( $10^{-5}$  mbar). Data of  $\eta$  values from mechanically cleaned and *in situ* Ar ion bombarded samples are compared with other published  $\eta$  data.

**Key Words**: Backscattered electron coefficients, low voltage microscopy, scanning electron microscopy.

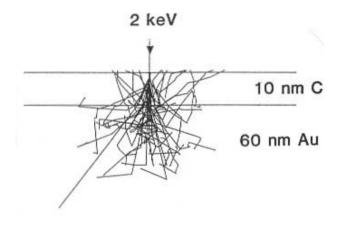
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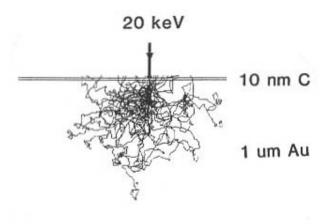
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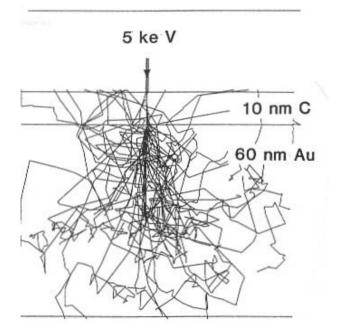
## Introduction

Low voltage scanning electron microscopy (LVSEM) is useful in studies where a decrease in the electron range, reduced charging and radiation damage, and an increased topographic contrast are desired. Such characteristics have seen an increased use of LVSEM in the semiconductor industry, in metrology and in biological science (Ogura, 1991; Perovic et al., 1995; Venables and Maher, 1996). LVSEM is therefore witnessing intensive activity in both instrument manufacture and image interpretation. The former is due to the recent developments in electron sources, where the higher brightness field electron emitters of the Schottky type are increasingly replacing conventional thermionic sources. A corollary of this development is that small probe diameters with sufficient probe current to form an SEM image of comparable signal to noise ratio to conventional large probe diameter SEM's are now commercially available (Ogura, 1991). Image interpretation on the other hand is still in its infancy despite a large data base of experimental data of the backscattering coefficient η (Bishop, 1967; Darlington and Cosslett, 1972; Fitting, 1974; Hunger and Kuchler, 1979; Reimer and Tollkamp, 1980; Bongeler et al., 1993) and predictions by Monte Carlo simulations (Ichimura and Shimizu, 1981; Joy, 1987; Bongeler et al., 1993).

The backscattering coefficient  $\eta$  is conventionally defined as the ratio of the number of electrons that backscatter out of the sample surface (I<sub>B</sub>), with an energy greater than 50 eV, to the total number of the incident electrons ( $I_p$ ), i.e.,  $\eta = I_R$ /  $I_{\rm p}$ . To our knowledge, most of the experimental results reported to date have been measured under conventional vacuum conditions (10<sup>-5</sup>-10<sup>-7</sup> mbar). Thomas and Pattinson (1970) and Darlington and Cosslett (1972) have reported backscattering measurements at 10<sup>-8</sup> mbar. However, none of these studies had any provisions for *in situ* surface cleaning. We believe that these reported  $\eta$  measurements are, therefore, most likely to be from samples which have a thin surface film of contaminants that is not representative of the underlying bulk properties. Such a film is likely to vary in thickness from one sample to another and greatly dependant on surface pretreatment. However, its effect is to alter the backscattering behaviour of the part of the sample under electron bombardment, particularly at these low electron energies.







**Figure 1**. A simulation of electron solid interaction for a film of carbon 10 nm thick on a gold substrate. A number of incident beam energies,  $E_p$ , are considered: (a) 2 keV (b) 5 keV, and (c) 20 keV. Note that as  $E_p$  increases more of the backscattered electrons travel into the Au substrate. This gives rise to increased  $\eta$  values in the above cases of 0.212, 0.44, and 0.5, respectively.

From electron-solid interaction simulations (Assa'd, 1996), it can be shown that 2 keV electrons impinging a solid surface at normal incidence will usually have a maximum range in the region of 30-80 nm depending on the target atomic number. Electrons of energy in excess of 5 keV will, however, have a range greater than 100 nm. Further, it is well known that most solid surfaces grow a natural layer of contamination when exposed to atmospheric conditions. This layer could extend to several nanometers and mainly consist of oxygen, carbon and hydrogen compounds. The backscattered electrons, on the other hand, travel on average to a maximum depth of only about a third of the primary electron range. At 2 keV, this will be about 10-30 nm. It is therefore obvious from

the above discussion that any measurement of  $\eta$  at energies less than 2 keV will reflect the presence of the surface film, particularly for higher atomic number materials.

Figure 1 depicts an illustration of this effect where a carbon layer of thickness 10 nm is deposited on a gold substrate. As the energy of the incident electrons is increased, η also increases from 0.212 for 2 keV electrons to 0.5 for 20 keV electrons. The model used in this simulation is based on that by Joy (1987) but with a modified Rutherford cross-section to give backscattering coefficients closer in value to those obtained experimentally at electron energies 2-30 keV (Assa'd et al., 1992). However, for the purpose of these illustrative simulations, the details in the scattering model used will not change the values to the magnitude experimentally observed.

It was with this picture in mind that we started the present study. In so doing, we adopted the design of the detector reported by Reimer and Tollkamp (1980) to a ultra high vacuum (UHV) environment.  $\eta$  values in the energy range 0.5-6 keV and at normal electron beam incidence have been measured for a number of elements before and after their surfaces were *in situ* cleaned with energetic ions.

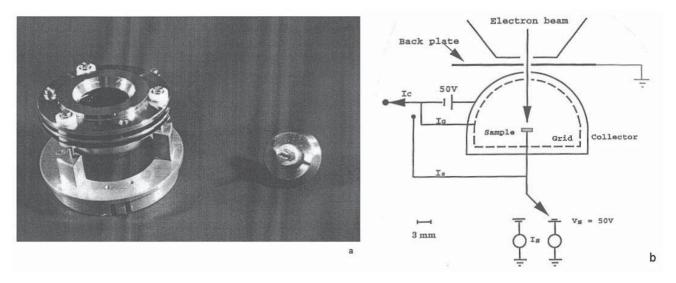


Figure 2. (a) The backscattered detector used for the measurement of  $\eta$ , (see text for details); (b) a schematic of the detector and the experimental set-up used in the present experiments.

# **Experiment**

The backscattered electron detector used in this work is shown in Figure 2. It is based on that reported by Reimer and Tollkamp (1980) but with two basic differences as follows:

(1) The collector electrode is made out of molybdenum in the shape of a hemisphere of radius 15 mm, with the sample placed at its centre, making a collection angle of  $2\pi str$ . The hemisphere extends 3 mm into a cylindrical shape and is connected to a 30 mm diameter plate that carries the sample. This ensures that all electrons that may backscatter from the collector are captured.

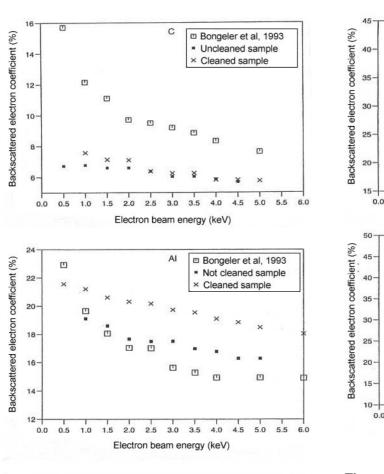
(2) The sample can be moved *in situ* from the backscattered detector to face an energetic ion beam for surface cleaning. In addition, the present design allows a carousel type sample manipulator to be used, and hence a number of samples can be investigated without the need to break the vacuum seal. This is a novel feature of the present detector.

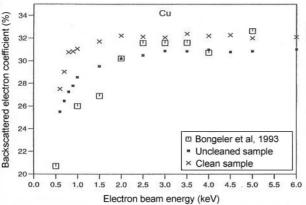
The grid used in the detector is made out of stainless steel with a wire gauge of 100 microns. It has 85% transparency. All the inner surfaces of the metal parts of the detector that are exposed to electrons as well as the grid electrode were coated with a low electron emission carbon compound (GRAPHIT 33, Kontakt Chemie, GMBH, D-7557, Iffezheim, Germany). The use of this material ensures that a minimum of electron emission (secondaries and tertiaries) occurs from the interaction of electrons emitted from the sample with the grid. Since the design of the present detector is similar in principle to that of Reimer and Tollkamp (1980); a figure of systematic errors in

the measurement of the backscattering and secondary electron coefficients as obtained by these authors, amounting to less than 1%, is therefore expected. An analysis of these sources is given by Reimer and Tollkamp and for the sake of brevity will not be repeated here.

The experimental setup used consisted of a vacuum system bakeable to 160°C to achieve a base pressure in the region of 2-3 x 10<sup>-10</sup> mbar, a variable energy 0.6-10 keV electron beam having a current in the range of 0.1-1 x 10-6 A in a spot of about 10-30 µm diameter. Ion beam cleaning conditions were 2-4 x 10<sup>-6</sup> A at 3 keV and typically were used for about 30 minutes. These ion beam conditions have been used previously in cleaning similarly prepared surfaces for Auger electron spectroscopy applications (El-Bakush and El-Gomati, 1995). The samples were made of thin foils of high purity materials (99.999%) and were typically 0.5 mm thick and 2-3 mm in diameter. The beam current stability was better than 0.02% per hour and all current measurements were made with a Keithley 604 electrometer (Keithley Instruments, Inc., Cleveland, OH). The measurements were repeated at least twice, and  $\eta$  values were found to be reproducible with a standard deviation of better than 1%.

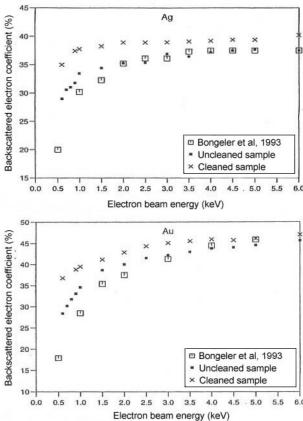
Backscattering coefficients  $\eta$  were measured in the energy range 0.6-6 keV for C, Al, Cu, Ag and Au at normal incidence. Figure 3 shows the results obtained for both the uncleaned (as inserted) and cleaned samples, as well as the experimental data of Bongeler *et al.* (1993). With the exception of carbon and aluminium, all three data sets show a similar pattern of low  $\eta$  value at low energies rising with different degrees of steepness as  $E_n$  is increased up to 3-4 keV where





the increase becomes smaller.

The backscattering values of the Ar $^+$  ion cleaned surfaces are plotted in Figure 3. It is clearly seen from these data that the  $\eta$  values obtained at low electron energy are higher than those for the uncleaned sample. In addition, it is also found that the  $\eta$  values of any element from the clean surfaces increase only slowly as a function of the incident beam energy with a maximum range of about 20% (for Au, at  $0.6 \, \text{keV}$ ,  $\eta = 0.37$ , and at  $6 \, \text{keV}$ ,  $\eta = 0.47$ ). This is in contrast to



**Figure 3**. The backscattered electron coefficient as a function of incident electron beam energy for (a) carbon, (b) aluminium, (c) copper, (d) silver, and (e) gold. Open symbols are from Bongeler *et al.* (1993); filled symbols and crosses are the "as inserted" and the ion cleaned samples, respectively, from the present work.

the uncleaned surface (as inserted) which shows an increase by more than 60% (for Au, this corresponds to  $\eta=0.17$  at 0.6 keV to  $\eta=0.46$  at 6 keV). These ratios assume that the  $\eta$  value at 6 keV is nearly constant for both the as inserted and cleaned surfaces. The behaviour for as inserted samples is in qualitative agreement with the discussions of Figure 1 describing a thin film of low Z material deposited on a substrate of a higher Z material. It is important to note that the film thickness affects the absolute value obtained for a given beam energy. We have confirmed this pattern by collecting  $\eta$  at various stages during the cleaning process of Cu and Au surfaces. While the  $\eta$  value at 5 keV remained approximately the same, the measured values at less than 1-2 keV increased as the cleaning

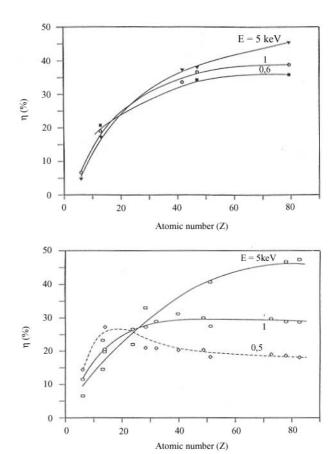
progressed.

In the case of the cleaned samples, the measured backscattered coefficients were found to reduce in value only slightly under prolonged electron beam bombardment. This, on the one hand confirms the cleanliness of the surfaces, while on the other, it shows that carbon deposition on target surfaces under electron bombardment is much reduced under UHV conditions.

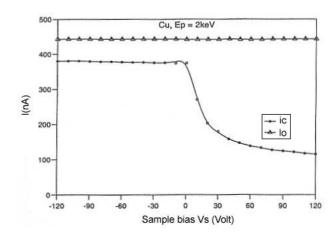
It is interesting to note that the as inserted data, which is expected to agree more with the data of Bongeler et al. (1993), in fact falls in between it and the data from the cleaned samples. This perhaps is because all three surfaces have different contamination levels and that also the experiments were carried out in different environments. Under UHV conditions, it is expected that carbon and oxygen deposition are reduced, and therefore the data of the as inserted samples of the present study will be slightly higher in value than the Bongeler et al. (1993) data, as mentioned above for the cleaned surfaces. An exception to this is carbon (and perhaps similar low atomic number materials). In this case, it is likely that a compound of average atomic number greater than carbon (e.g., containing oxygen) is formed on the Bongeler et al. (1993) sample to give a high  $\eta$  value at low  $E_p$ . For the as inserted samples in the present study, on the other hand, it is likely that a compound of lower average atomic number than carbon (e.g., containing hydrogen) has formed on its surface to give a low  $\eta$  value at low  $E_n$ .

Figure 4 depicts the relationship between  $\eta$  and the target atomic number (Z) at different incident beam energies for the clean samples (Fig. 4a) and the as inserted samples (Fig. 4b), respectively. The data for the as inserted samples are those of Bongeler *et al.* (1993). It is clearly seen that for the cleaned samples  $\eta$  increases as Z is increased for all energies. In the case of the as inserted samples, the relationship between  $\eta$  and Z for low beam energies is different than those at higher energies. For  $E_p = 0.5$  keV, low Z materials give higher  $\eta$  values than higher Z materials, while for  $E_p = 5$  keV,  $\eta$  increases as Z increases. In addition, for Z > 30 and  $E_p = 0.5$  keV,  $\eta$  is constant to within  $\pm 2\%$ . In contrast, the clean samples show a systematic increase in  $\eta$  for all energies used.

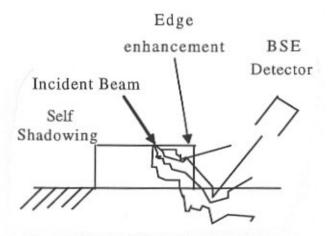
The results shown in Figure 4a are in contradiction to previously published experimental data, and although the range of samples investigated here is limited, the results obtained show a pattern suggesting that unambiguous atomic number contrast reflecting high  $\eta$  values for high atomic numbers can also be obtained in low as in high voltage microscopy. The price to be paid for such a restoration of atomic number contrast is the provision of  $in\ situ$  sample cleaning and vacuum conditions that preserves surface cleanliness during the measurement, as in UHV surface analysis. This may not be easily achieved in most SEM's which employ conventional vacuum. However, with the ever increasing use of field electron emitter sources in SEM and



**Figure 4**. The backscattered electron coefficient as a function of the atomic number for different electron beam energies (**a**) for Ar ion cleaned surfaces, (**b**) as inserted from Bongeler *et al.* (1993).



**Figure 5**. Retarding the field plot of current versus specimen voltage collected from a Cu sample at 2 keV incident electrons.



**Figure 6**. Schematic illustration of the effect of topography on the measurement of the backscattering coefficient. Both shadowing and edge enhancement are functions of the detector collection angle.

their operational requirement of UHV environment, manufacturers and prospective customers of future high resolution low voltage SEMs should perhaps take such a practice into consideration.

# Conclusion

New experimental data of the backscattering coefficients of C, Al, Cu, Ag and Au at normal angle of incidence and low electron energies (0.6-6 keV) are reported. The results obtained are from samples that have been cleaned in situ with energetic ions to remove any surface contamination prior to measurements. The  $\eta$  data are collected under UHV conditions to preserve surface cleanliness. An electron detector, for the measurement of the backscattering coefficient, with provision for in situ sample cleaning is also reported. In contrast to current data in the literature, the present results at low electron beam energies (less than 2 keV) show a monotonic increase of  $\eta$  with increasing target atomic number. It is our opinion that the previously reported data are likely to be from samples which contain a thin, but significant, surface film of low atomic number contaminants that may have arisen as a result of sample preparation or experimental conditions. At low incident electron energies, it is this film that dominates the interaction volume of the electron with the solid, giving lower backscattering coefficients than expected from cleaned surfaces. This may also be responsible for the reversed contrast between low and high energy imaging as reported by Ogura (1991).

Work is in progress to collect data from additional

materials spanning the periodic table and at other incident angles. In addition, the secondary electron coefficients of the same targets are also being measured. A theory/experiment comparison of these data involving Monte Carlo simulations is in progress and will be reported shortly.

## Acknowledgements

The authors would like to thank Professor J.A.D. Matthew and Dr. S.J. Bean (York Electron Optics Ltd., University of York) for critical reading of the manuscript and Mr. Iain Will for fabricating the detector. This work has been partly supported by EPSRC and CEC grant no. CP 12283.

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#### **Discussion with Reviewers**

**M. Dapor**: The number of trapped electrons in the contamination film and that of electrons transmitted through it should influence the backscattering coefficient measurement. The transmission and absorption of electrons through the film of contaminants depend, for a given thickness, on the electron primary energy and on the kind of contaminants. Can the authors give an evaluation of the fraction of electrons trapped and of that of electrons transmitted for a typical layer of contaminants (carbon, for example) and for low energy ( $E_n < 1000 \, \text{eV}$ ) electrons?

Authors: We have recently developed a fast Monte Carlo code for the generation of all the signals used in multi-spectral Auger microscopy (MULSAM); namely, Auger, secondary and backscattered electrons and X rays. It is based on a modified Rutherford scattering cross-section. The modified formula compares well with the more exact Mott cross-section, particularly at low electron energies and high atomic number materials. Details of this model will be submitted later to this journal. Using this model and simulating an incident electron beam on a film of carbon deposited over targets of different atomic numbers ranging from carbon to gold shows that at least between 60-70% of the incident electrons are trapped within the carbon layer, depending on the target material.

**H.J. Fitting**: I fear an affection of  $\eta = I_{c+}/I_{p}$  by means of "tertiary" electrons emitted from the mesh stages back to the sample. It may be the reason that I find your data generally lower than our data of a comprehensive review in H.J. Fitting et al. (1991) Phys. Stat. Solidi (a) **126**, 8500. This systematic error should be determined and avoided as much as possible. **Authors**: The measurement of  $\eta$  is carried out as follows: I is the current measured by the collector and grid with the sample positively biased to 50 V. In this case, any secondaries or tertiaries emitted in the system will be collected by the positively biased sample. The primary beam current I is measured when the sample, grid and collector are all connected together.  $\zeta$  is then calculated as  $\eta = I_{\alpha}/I_{\alpha}$ . Reimer and Tollkamp (1980) have discussed and estimated the likely errors in the measurement of  $\eta$  that could result from secondary and tertiary electrons which are generated by backscattered electrons

(BSE) at the grid and collector and collected by the positively biased sample. In the present setup, which is similar in principle to that by these authors and for a sample of no more than 3 mm in diameter, and a carbon coated grid, this has been estimated at less than 0.3% of the backscattered coefficient (see Reimer and Tollkamp, 1980, for further details).

The sample bias used here is determined after a measurement of a retarding field plot of the current  $I_c$  versus sample bias. The plot for Cu at 2 keV is shown in Figure 5. Careful inspection of Figure 5 shows that  $I_c$  is still decreasing as the sample bias is increased. The difference between 40 V and 50 V biasing amounts to a decrease in  $\eta$  of just under 3% in this case and for the samples studied here is found to be in the range of 2-3%, which is similar in value to the calculations of Fitting *et al.* (1991). However, we chose to use 50 V biasing to distinguish between secondary and backscattered electrons for consistency with well established practice in the literature.

**Z. Radzimiski**: What kind of precleaning did you use before inserting samples into the vacuum chamber? Can you identify a cleaning method which would be most suitable for sample preparation?

Authors: All samples were degreased in deionised water followed by an ultrasonic bath in isopropanol alcohol for about 5-10 minutes, and then were dried before being inserted into the vacuum system. This is the method normally used in our surface analysis systems. Its main function is to ensure that no residual foreign materials are left on the surfaces, particularly grease from handling the samples. This also helps one obtain a true UHV environment in a reasonable time, normally between 12-24 hours at 160°C. In the case of conventional SEM environment, the use of a similar procedure should eliminate any presence of foreign materials on the surface. However, even with this cleaning procedure, the experiments performed here show that in both cases, and with the use of low energy electrons, one would still need the use of UHV and the *in situ* sample cleaning to obtain the true  $\eta$ and  $\delta$  values of the constituent elements.

**Z. Radzimiski**: Could you give some evaluation of theory/experiment comparison. You have presented Monte Carlo data in this paper, so I assume that you have already some theoretical values of BSE coefficients.

**Authors**: The data presented in this study are all experimental measurements. Preliminary results from the MULSAM Monte Carlo code mentioned above indicate a similar pattern to the data presented here up to about 1 keV. However, for electron energies below this value, the level of agreement is considerably less than at higher energies, and this is being currently investigated, particularly with respect to electron energy loss expressions.

K. Murata: Surface roughness is also an important factor for

backscattering at low energies. Could you comment on how the roughness influence the backscattering coefficient? How did you confirm flat surfaces experimentally?

Authors: The sample roughness is indeed an important factor to be taken into consideration in the measurement of the backscattering coefficient from a solid target. This is because the position of a topographical feature on the surface with respect to that of the incident electrons, on the one hand, and the electron detector and its angle of collection on the other hand, will give rise to two artefacts. These are self-shadowing and edge enhancement as depicted in Figure 6. However, these artefacts are normally seen in the case of an incident electron beam of diameter much less than the height of the feature being imaged. Its effect, however, will still be present in the total yield, and therefore, flat surfaces must be used in these type of measurements. Another important factor is that backscattering normally increases as a function of the incidence angle, hence for topographically rough surfaces,  $\eta$ is expected to increase depending on the degree of surface roughness.

In the present study, the samples were all mechanically polished to about two microns. This was established optically before insertion into the vacuum chamber and by using the sample imaging facility of the Varian Auger spectrometer used here, which has a resolution of  $10\text{-}20~\mu m$  at these energies.

**D. Venables**: Could the authors please elaborate on why the low Z materials show an increase in  $\eta$  as the beam energy decreases, whereas high Z materials show the opposite trend? **Authors**: The behaviour of  $\eta$  for electrons of energy less than 2 keV is believed to be largely affected by the ratio of elastic to inelastic scattering of electrons with the target material. Inelastic interaction is a function of a number of losses; plasmon excitation, band transition and electronelectron collision, all is believed to be strongly target dependant in a complex way that does not reflect a simple linear atomic number dependence as the case is for higher electron energies. However, in our opinion, there may be some systematics of  $\eta$ across the periodic table at low incident electron energies that give rise to such losses in the first place. Such systematics could only be discovered with data obtained from clean sample surfaces, and the present work is a step towards this goal.

**D. Venables**: The authors suggest that the differences between their own as-inserted data and that of Bongeler *et al.* (1993) may be due to the difference in vacuum conditions under which the data was acquired. In this regard, how do the data of Darlington and Cosslett (1972) (also acquired under UHV conditions) compare to those of the authors and to those of Bongeler *et al.* (1993)?

**Authors**: The data of Darlington and Cosslett (1972) were indeed obtained under UHV conditions but there was no *in situ* surface cleaning facilities used. In this regard, it could be

that the surfaces studied may have a layer of natural contamination of unknown thickness and composition. In the authors experience with surface analysis, there will always be an element of contamination on solid surfaces even on surfaces which have been degreased in a solvent. In the case of the data of Bongeler et al. (1993), in addition to the natural contamination layer, the data was collected under conventional vacuum conditions in a commercial electron microscope. Poor vacuum conditions could cause an additional layer of adsorbates on the sample surface. Both sets of data are therefore different than those presented here, with the data of Bongeler et al. (1993) more likely to show a stronger contamination than that of Darlington and Cosslett (1972). The data presented by Darlington and Cosslett (1972) generally shows higher values than either our data or those by Bongeler et al. (1993).