Supplementary Information

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Appendix

A Database of Electron–Solid Interactions

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A Database of Electron–Solid Interactions

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Abstract
A collection of data comprising secondary and backscattered electron yields, measurements of electron stopping powers, and X-ray ionization cross sections has been assembled from published sources. Values are provided for both elements and many compounds, although the quality and quantity of the available data vary widely from one material to another. These compilations provide the basic framework for understanding and interpreting electron beam images in a quantitative way—as is required for example in semiconductor device metrology—and also form a comprehensive source of experimental data for testing analytical and Monte Carlo models of electron beam interactions.

Introduction

The year 1997 marked the one hundredth anniversary of the discovery of the electron. Within a year of that event Starke (1898) in Germany, and Campbell-Swinton (1899) in England, independently showed that electrons were backscattered from solid specimens and so made the first quantitative measurements of the interaction of electrons with material. Over the century since then many dozens of papers have been published that contain information on various aspects of electron–solid interactions. Unfortunately no systematic collections of the results of such investigations appear to be available for any part of the field of electron microscopy and microanalysis. As a result, anyone requiring a specific piece of data—such as the backscattering yield of molybdenum at 15 keV, or the secondary electron yield from gallium arsenide at 3 keV to take two random examples—has no option but to search the literature in the hope of finding a value which must then, in the absence of any other comparable evidence, be taken as correct. What is required is a source which collects and collates all the values available so as to provide the user with not only a value, but some indication as to its likely reliability.

Structure of the Database

The database presented here is an attempt to present as complete a survey as possible of the published results on backscattering yields, secondary electron yields, stopping powers, X-ray ionization cross sections, and fluorescent yields. Computer-aided literature searches have been conducted to try and find all published references in this general area for the period from 1898 to the present day. Clearly no claim can be made as to the completeness of any such search, and it is perhaps to be hoped that some major body of work has been overlooked because, as discussed below, there are otherwise major omissions in the materials available.

The rules for the data included in this collection are simple:

(a) Only experimental results are included. Values that are not specifically indicated by the author(s) as being experimental, or values that are clearly the result of interpolation, extrapolation, or curve fitting, have been expunged.
(b) No attempt has been made to critically assess the accuracy or precision of the data, nor to remove any results on the basis of their presumed quality.
(c) Values have been tabulated primarily for the energy range up to 30 keV, although data points for incident energies up to 100 keV have been included where they are available.

The decision not to engage in any judgment of the quality of any of the sets of results may seem to be a significant drawback to the utility of the database. However, until so much data has been collated for each element or compound that rogue values can infallibly be distinguished and eliminated, there is no criterion on which to reject any particular result. Further it is conceivable that two tabulated values of a given parameter may differ substantially and yet still both be of value. This is because of an inherent contradiction in the nature of the measurements that are being made. A measurement made in a UHV electron scattering machine with *in situ* sample cleaning and baking facilities will naturally be more “reliable” than a measurement made inside a typical scanning electron microscope. But the values recorded in the microscope are more “representative” of the conditions usually employed on a day-to-day basis in an e-beam tool than those obtained in the environment of a specialist instrument. All types of results are, therefore, reported so that users of the database can make their own judgment as to the suitability, or otherwise, of any given piece of information.

The database currently contains several thousand individual values collected from more than 100 published papers and reports spanning the period from 1898 to the present day. Since this is a "work in progress" the compilation is constantly being extended as additional values become available. As far as possible a consistent style of presentation is used so that data for different elements and compounds may readily be compared. All of the available data sets are grouped by element or compound name for each of the major information groups (SE yields, BSE yields, stopping powers, X-ray ionization cross sections). The data is presented in a simple two-column format with the origin of each of the data sets (numbered #1 to #n) indicated by a number in parenthesis referring back to the bibliography.

Backscattered Electrons

The data on the backscattered electron (BSE) yield as a function of the atomic number of the target and of the incident beam energy is of particular importance in Monte Carlo computations because it provides the best test of the scattering models that are used in the simulation. This data is therefore both the starting point for the construction of a Monte Carlo model, and the source of values against which the simulation can be tested. The backscattered electron section contains data for 40 or more elements spread across the periodic table, as well as for a selection of compounds. If Castaing's rule can be assumed to be correct, then the backscattering yield of a compound can be found if the backscattering coefficients and the atomic fraction of the elements that form it are known. Hence a desirable long-term goal is to obtain a complete set of BSE yield curves for elements. At present the BSE section contains information on more than 45 elements, which is barely half of the solid elements in the periodic table, and of this number perhaps only 25 % of the data sets are of the highest quality, so much experimental work remains to be done especially at the lower energies.

Secondary Yields

With the increasing interest in the simulation of secondary electron (SE) line profiles and images, there is a need to have detailed information on secondary electron yields as a function of atomic number and incident beam energy. Secondary electron emission was the subject of intense experimental study for a period of 20 years or more from the early 1930s, resulting in the publication of no less than six full-length books on the topic. This effort did not, however, produce as much experimental data as would have been expected, because the aim of much of the work that was done was to demonstrate that the SE yield versus energy curve followed a "universal law" (Seiler 1984), and to find the parameters describing this curve. As a result the data actually published was usually given in a normalized format that makes it difficult to derive absolute values. The database currently contains yields for about 40 or elements, and a collection of inorganic compounds and polymers.

The clear discrepancies that often exist between the comparable sets of original yield figures for the same material may be the result of surface contamination, or the result of a different assumption about the appropriate emitted energy range for secondary electrons (usually now taken to be 0–50 eV, although in some early work 0–70 or even 0–100 eV was used). In addition, since many of the materials documented are poorly conducting the effects of charging must also be considered. For example, in studies of the oxides (e.g., Whetten and Laponsky 1957) maximum SE yields of $\delta > 10$ were measured using pulsed electron-beam techniques. Clearly no non-conducting material can sustain this level of emission for any significant period of time, since it will become positively charged and recollect its own secondaries. Similarly at higher energies, where the SE yield $\delta < 1$ and negative charging occurs, the incident beam energy must be corrected for any negative surface potential acquired by the sample to give a correct result (although there is no little evidence in the original papers that this has been done). Consequently, all SE yield results for insulators must be treated with caution unless the provenance of the original data is well documented.

Since there is no sum rule for secondary yields, data must be acquired for every compound of interest over the energy range required, a task which will be a lengthy one unless suitably automated procedures can be developed and applied. In addition it will be necessary to repeat many of the measurements reported here using better techniques before any level of precision and accuracy can be obtained. In summary the SE data is in an even less satisfactory state than that for the BS electrons, even though a wider range of materials is covered, because the quality of much of the data is poor.

Stopping Powers

The stopping power of an electron in a solid, that is, the rate at which the electron transfers its energy to the material through which it is passing, is a quantity of the highest importance for all studies of electron-solid interactions since it determines, among other parameters the electron range (Bethe 1930), the rate of secondary electron production (Bethe 1941), the lateral distribution and the distribution in depth of X-ray production, and the generation and distribution of electron–hole pairs. Despite its importance there is no body of experimental measurements of stopping power at those energies of interest to electron microscopy and microanalysis. Instead stopping powers, and the quantities which depend on them, have been deduced by analyzing measurements of the transmission energy spectrum of MeV-energy β-particles to yield a value for the mean ionization potential I of the specimen (ICRU 1983), and then Bethe's (1930) analytical expression for the stopping power has been invoked to compute the stopping power at the energy of interest. While this procedure is of acceptable accuracy at high energies (>10 keV) it is not reliable at lower energies because some of the interactions included in the value of I (e.g., inner shell ionizations) no longer contribute.

The database contains experimentally determined stopping power curves for a collection of elements and compounds. The method for obtaining this information from electron energy loss spectra has been described elsewhere (Luo et al. 1991). The data is plotted in units of eV/Å as a function of the incident energy in keV. At the high energy end of the profiles the data corresponds closely to values deduced from Bethe's (1930) law and using the I-values from the ICRU tables. At lower energies, however, significant deviations occur as the Bethe model becomes physically unrealistic although good agreement has been found with values computed from a dielectric model of the solids (Ashley et al. 1979).

The stopping power of a compound is the weighted sum of the stopping power of its constituents; thus a key priority for future work should be to complete the set of stopping power profiles for elements rather than to acquire more data on compounds.

X-ray Ionization Cross Sections

Measured values of the X-ray ionization cross sections for various elements and emission lines as a function of incident beam energy are also of great importance in microanalysis. Unfortunately, as a brief study of the graphs included here will show, the amount of data available is small for K-shells, negligible for the L-shells, and all but non-existent for the M-shells and higher. This is the result of pervasive experimental difficulties, in particular, the fact that any measurement couples together the ionization cross section and the fluorescent yield ω. Since, as can be seen from the plots in section 5 of the database, the value of the fluorescent yield ω is poorly known for the L- and M-shells this causes a significant degree in uncertainty in the cross section deduced from this data. A more practically useful approach is, instead, to quote an "X-ray generation" cross section which is the product of the ionization cross section and the fluorescent yield term. Because the fluorescent term is never required separately in X-ray microanalysis this result looses nothing of its generality but is much more robust. Future updates of this database will include results in this format. For completeness section 5 tabulates all the available fluorescent yield data for K-, L-, and M-shells.

Conclusions

This database is a first step toward the goal of providing a comprehensive collection of the parameters which describe electron–solid interactions. In addition to meeting the needs of those working in Monte Carlo modeling, it is hoped that a systematic collection of data such as this may also be of value in experimental electron microscopy. The quality and quantity of the data that has been amassed varies widely from one material, and from one topic, to another, so that while a few elements can be considered as well characterized, the overall situation is poor, especially for materials used in such areas as integrated circuit device fabrication.

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