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A Database on Electron-Solid Interactions

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Summary: Monte Carlo modeling of electron-solid interactions requires a detailed and accurate supply of experimental data on which to base its physics and against which to test its predictions. To meet this need, a collection of data—comprising measurements of secondary and backscattered electron yields, electron stopping powers, and x-ray ionization cross sections, as a function of energy—has been assembled from published sources. The quality and quantity of the compilation varies widely, with little or no data being available for the majority of elements in the periodic table, while results for complex materials of current technologic interest are also almost nonexistent. To meet the needs of Monte Carlo simulation in areas such as dimensional metrology or microanalysis, a program of systematic measurements is required.

Key words: secondary electron yield, backscattered electron yield, ionization cross section, stopping power

Introduction

A Monte Carlo simulation is only as good as the experimental data which went into setting it up and against which it was subsequently tested. The continued development and increasing sophistication of Monte Carlo techniques for simulating electron-solid interactions will therefore necessarily result in the call for more detailed and comprehensive sets of relevant data to be available to the researcher in an accurate and convenient form. This paper describes progress on an attempt to produce a database containing the type of information on electron scattering that is required.

The first quantitative measurements of the interaction of electrons with material were examinations of the backscattering yield from solid specimens made independently by Starke (1898) in Germany, and by Campbell-Swinton (1899) in England. 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 such data appear to be available for any part of the field of electron microscopy and microanalysis. As a result, anyone requiring specific information such as the backscattering yield of molybdenum at 15 keV, or the secondary electron yield from GaAs at 3 keV, to take two arbitrary examples, has no option but to search the literature randomly in the hope of finding a quoted value which must then be taken as reliable in the absence of any other comparable evidence. This is an undesirable situation in comparison with that found, for example, in Proton-Induced X-ray Analysis (PIXE) for which there is a computerized collection of data covering all aspects of the proton-solid interaction, which has been compiled by a consensus of all competent workers in the field.

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Structure of the Database

The database presented here is an attempt to collect and correlate as many as possible of the published measurements of backscattering yields, secondary electron yields, stopping powers, and x-ray ionization cross sections. While other types of data would also be of general value to electron microscopists, for example, electron-hole pair energies or mean inner potentials, the amount of published material so far collected is insufficient to warrant such an effort at this point. Computer-aided literature searches have been conducted to find all citable, 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 such a search, and indeed it is 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 assess critically 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 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 integrity of the database. However, until sufficient data have been collated for each element or compound to allow obvious rogue values to be eliminated, there is no a priori basis on which to reject any particular result. Furthermore, it is conceivable that two tabulated values of a given parameter may differ substantially and yet still both be worthy of consideration. 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. However, the value recorded in the microscope is much more representative of the conditions usually employed on a day-to-day basis than the value 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 70 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 publications are retrieved. A consistent style of presentation is used as far as possible so that data for different elements may be readily compared. The current format of the database is as a collection of ASCII files for general purposes, or as CRICKETGRAPH™ files for the Apple Macintosh. There is one file for each element or compound, and these files are grouped together into subdirectories by the type of data contained in the files, that is, secondary electron yields, backscattered electron yields, x-ray ionization cross sections, and electron stopping power. Each file is formated to arrange the data in pairs of columns. The first column is always identified by a title of the form E(N) where E indicates the energy values (in keV) for the subsequent data, and the number N, for example 37, indicates that the source of the data was from reference number 37 in the bibliography. The second column contains one of four types of data identified by their title—SE for secondary electron yield, BS for backscattered electron yield, SP for stopping power (in eV per angstrom), or SG for x-ray ionization cross sections (in barns, where 1 barn = 10^{-24} cm^2/atom). The title also includes the reference number, for example, SE(37), to link it with the corresponding energy value column. The files can be read into most spreadsheet programs for further analysis and for plotting purposes. No attempt has been made in the data sets to indicate probable error limits, even in the (very few) cases where the original authors made such an estimate, because the best evidence of the accuracy is to be found from comparing the whole corpus of data for each element.

Backscattered Electrons

Backscattered electrons (BSEs) were the first effect of electron bombardment to be studied. The data on 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 this provides the best test of the scattering models that are used in the simulation. These data are 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 BSE section contains data for a variety of elements spread across the periodic table, as well as for a few compounds. Figures 1, 2, and 3 illustrate the levels in the quality of data that are available. Figure 1 shows the data set for gold, comprising values from nine authors in works published over a period of 30 years. The various values correlate excellently, and when taken together they measure the backscattering yield over the energy range from 1 to 100 keV with a scatter of only about ± 5%. Unfortunately, data of this quality and quantity are only found for a few other elements, such as Mo and Pt. Figure 2, which shows the data for copper, demonstrates a more common situation. Again, there is a substantial number of values in the literature which contribute to the composite curve, but the agreement between these values is poor, with discrepancies of nearly a factor of two between the minimum to maximum yields reported at some energies. Since the measurement of BSE yields is not inherently challenging, and since copper is neither so chemically active nor so impure as to present problems in sample preparation, it is not easy to see why this scatter exists, but this situation is found for many other elements such as C, Al, Si, and Cu. Finally, as in Figure 3 illustrating the data for Zn, most elements are only represented by one or two references and a sparse number of data points, generally providing insufficient data to allow any reliable or quantitative conclusions to be drawn.

Though the BSE section contains information on more than 40 elements, this is barely half of the solid elements in the periodic table, and of this number only perhaps 25% are of the quality of that shown in Figures 1 and 2. Values for compounds, even of the simplest and most common kind, are very scarce and only three partial sets, each with just one reference, seem to be available. Consequently, while these compilations are a great advance compared with the use of values drawn just from a single published report, the paucity and poor precision of the values that are included show that much work needs to be done. For example, it is impossible from the included data to answer even such a basic question as to whether or not the variation of backscatter yield with atomic number at a given energy is monotonic, or whether there are breaks in the yield curve at some atomic numbers as proposed by Bishop (1966). Similarly, the absence of data for compounds makes it impossible to verify whether or not Castaing's rule—that the BSE yield of a compound is the weighted sum of the backscatter yields of its constituents—is generally correct or not.

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 SE yields as a function of atomic

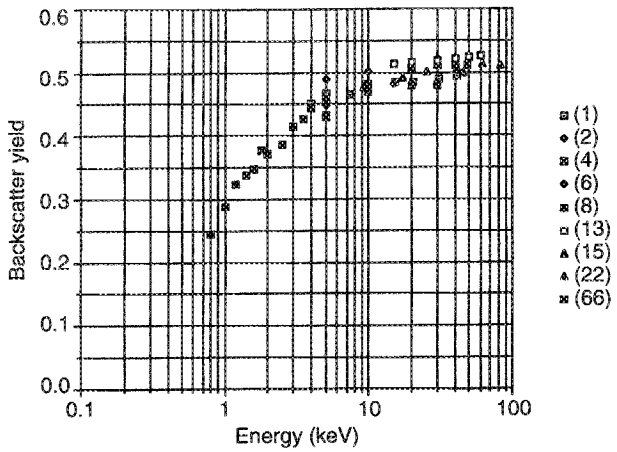

FIG. 1 Variation of backscattering yield for Au as a function of energy with data from nine authors. The numbers next to symbols correspond to references in the database bibliography.

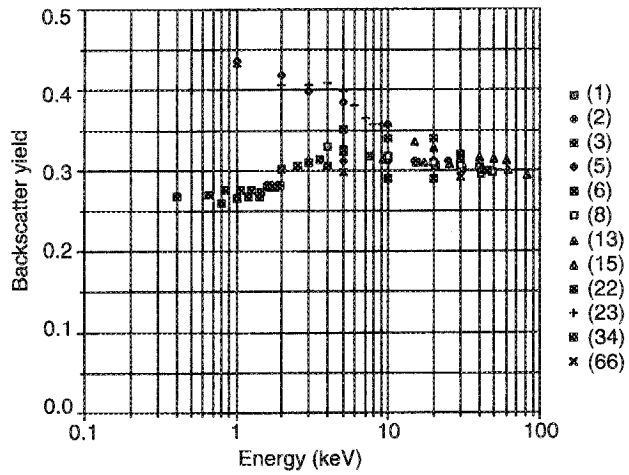

FIG. 2 Variation of backscattering yield for Cu as a function of energy with data from 12 authors. The numbers next to symbols correspond to references in the database bibliography.

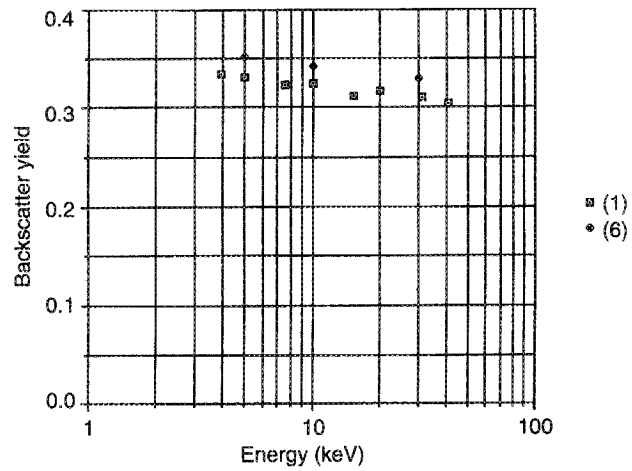

FIG. 3 Variation of backscattering yield for Zn as a function of energy with data from two authors. The numbers next to symbols correspond to references in the database bibliography.

number and incident beam energy. SE 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 useful experimental data as would have been expected, because the aim of most 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 defining this curve. As a result, the data actually published were usually given in a normalized format that makes it difficult to derive absolute values. The database currently contains yields for about 40 elements, and 20 or so inorganic compounds and polymers. Figure 4 shows an example of the best quality of the compilations available, in this case for silver, covering the incident energy range from < 100 eV to > 30 keV, and with generally good agreement between the multiple data sets. Regrettably, the corresponding collections available for most other elements have neither so many sets of points nor such good agreement, and more typically have the appearance shown in Figure 5 for silicon. The clear discrepancies between the various original yield values may be the result of surface contamination during the measurement, or the result of the authors making a different assumption about the appropriate emitted energy range for SEs (usually now taken to be 0 to 50 eV, although in some early work 0 to 30 or even 0 to 100 eV was used). In either case, the very wide spread of values makes it difficult to argue convincingly for the reliability of any particular point.

SE yield data have also been collected for some inorganic salts and for a few polymers. For example, Figure 6 shows data for the commercial polymer Kapton which has been studied by four separate groups, resulting in a yield curve with a modest level of apparent reproducibility. Most often only one or two sets of data points are available for such materials, and hence the precision and accuracy are in doubt. In addition, since these materials 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 $\partial > 10$ were measured using pulsed electron-beam techniques. Clearly, no nonconducting 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 $\partial < 1$ and negative charging occurs, the incident beam energy must be corrected for any 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 details of the original measurement protocol are well documented.

In summary, the SE compilation is in an even less satisfactory state than that for the BSE data, even though a wider range of materials is covered, because the scatter in yield values is high. It will be necessary to repeat many of these measurements using better techniques before any level of precision and accuracy can be obtained.

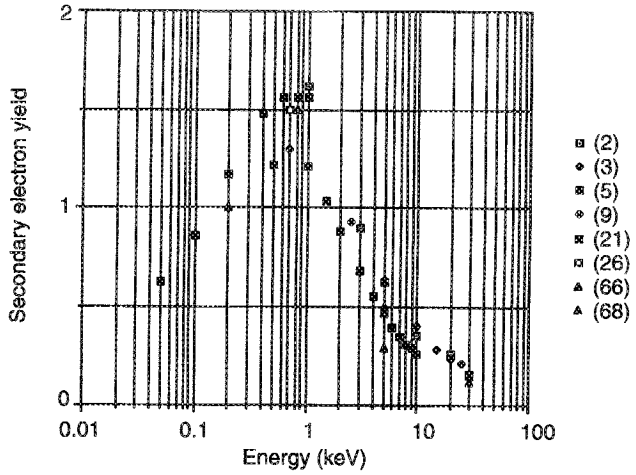

FIG. 4 Variation of secondary electron yield for Ag as a function of energy with data from eight authors. The numbers next to symbols correspond to references in the database bibliography.

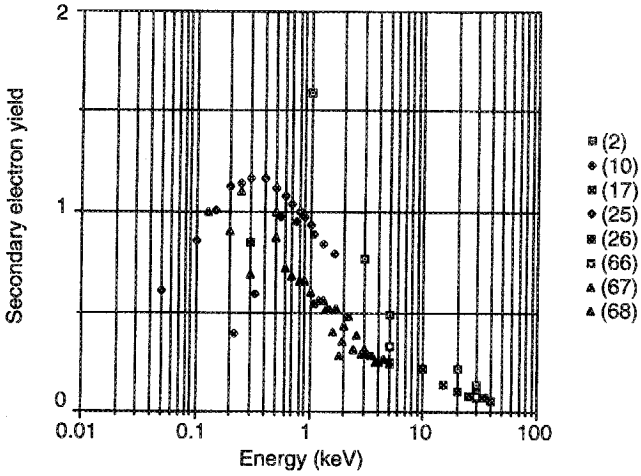

FIG. 5 Variation of secondary electron yield for Si as a function of energy with data from eight authors. The numbers next to symbols correspond to references in the database bibliography.

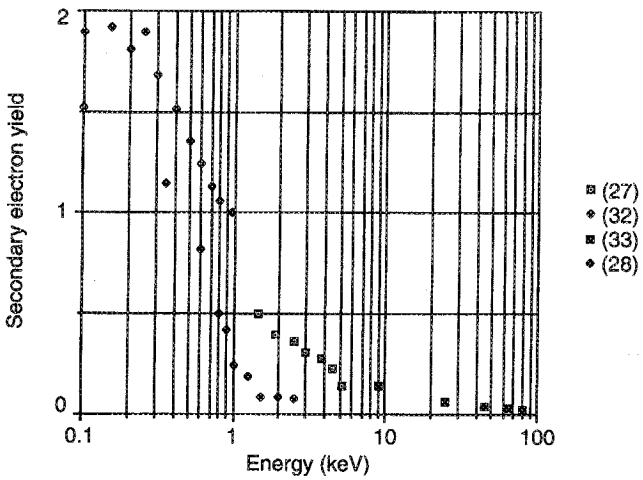

FIG. 6 Variation of secondary yield for Kapton as a function of energy with data from four authors. The numbers next to symbols correspond to references in the database bibliography.

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 ß particles to yield a value for the mean ionization potential I of the specimen (ICRU 1983) and then, using Bethe's (1930) analytical expression for the stopping power, to compute a value for the energy of interest. While this procedure is of acceptable accuracy at high energies (>10 keV), it is of dubious value at lower energies because some of the interactions included in the value of I (e.g., inner shell ionizations) are no longer available.

The database contains experimentally determined stopping power curves for a dozen elements and compounds. The method for obtaining this information from electron energy loss spectra has been described elsewhere (Luo *et al.* 1991). Figures 7 and 8 show the stopping power for carbon and for GaAs, respectively, plotted in units of eV/Å against the energy in keV. At the high energy end of the profiles, the data correspond 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). Since these data points are at present unique, they should be treated with the same reserve as other unsupported results until further independent, corroborative values are produced. In the meantime, these experimental stopping power curves indicate that the use of simplistic analytical models can lead to significant errors in Monte Carlo modeling, for example, in the computation of electron range, and therefore those experimental data should be used wherever they are available.

X-Ray Ionization Cross Sections

The final section of the database contains determinations of x-ray ionization cross sections as a function of incident electron energy. In this case, the amount of experimental data available is very limited as the majority of determinations have been made on a relative rather than an absolute basis. Data of good quality have been published for the K-shell excitations of low atomic number solids and gases. For example, Figure 9 illustrates the measurements for oxygen, from four authors, showing closely clustered values over a good range of ener-

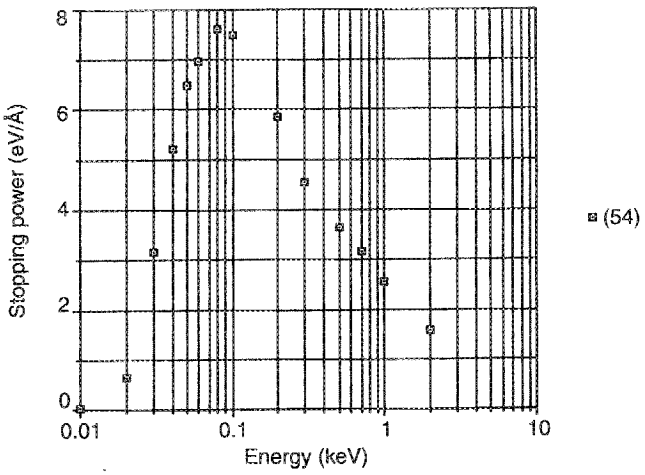

FIG. 7 Variation of stopping power for C as a function of energy. The number next to symbol corresponds to the reference in the database bibliography.

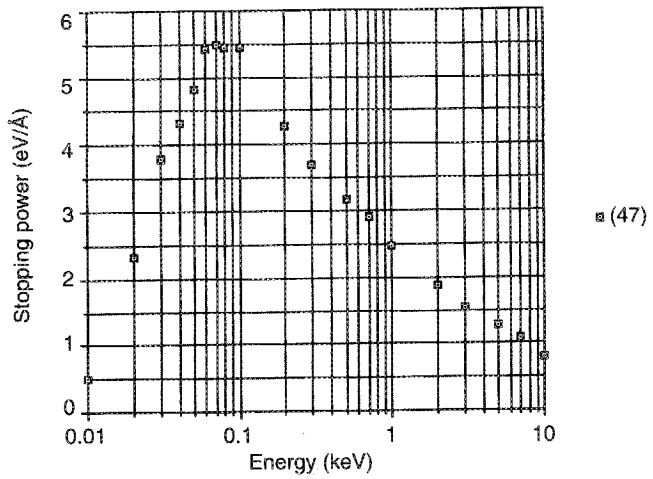

FIG. 8 Variation of stopping power for GaAs as a function of energy. The number next to symbol corresponds to the reference in the database bibliography.

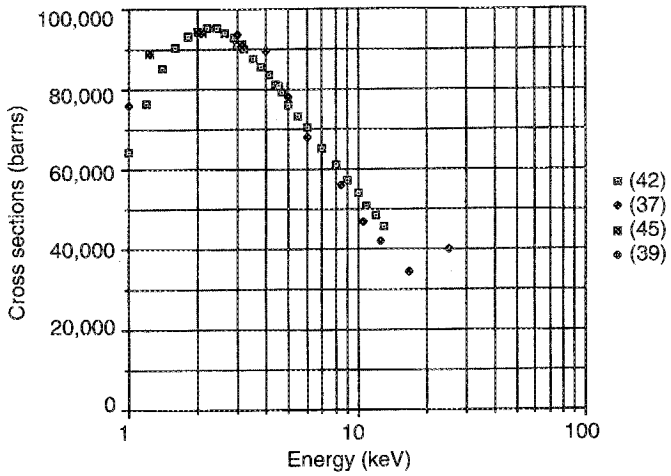

FIG. 9 Variation of ionization cross section for Carbon K-shell as a function of energy. The numbers next to symbols correspond to references in the database bibliography.

gies. For excitations other than K-shell , however, the corpus of published results is very sparse. Figure 10 illustrates data for the LIII excitation of gold. The very poor level of agreement between the four cited references indicates the difficulty of establishing a reliable value in this kind of situation. The major problem for absolute x-ray measurements is that the relevant fluorescent yield ω must be known. For K-shells, values of ω employed by different models are quite close and so contribute little error to the cross-section determination. However, for L, M, and higher shells there is little agreement as to the magnitude of fluorescent yields and this is reflected in the wide spread of cross-section determinations between comparable data sets. Since cross sections and the fluorescent yield invariably appear in an equation together, it might be preferable to quote the result in the form of their product to enable comparisons to be made more easily.

Conclusions

The quantitative modeling of electron-solid interactions using Monte Carlo techniques is only possible if there is sufficient experimental data on which to base the model and against which to test its predictions. This database is a first step toward that goal. In addition to meeting the needs of those working in Monte Carlo modeling, it is hoped that a systematic collection of data such as these may also be of value in answering other fundamental questions about electron-solid interactions. The quality and quantity of the data that have been amassed varies widely from element to element and from one type of measurement to another. While a few materials can be considered as being well characterized with regard to their properties under electron-beam irradiation, the situation overall is poor. For a majority of the materials likely to be encountered in current use, especially in areas such as integrated circuit device fabrication or materials research, no data are available at all. It is therefore to be hoped that the many and serious gaps in this compilation

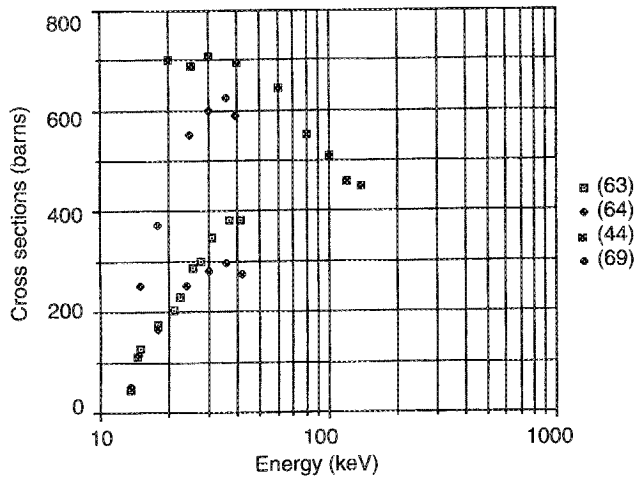

FIG. 10 Variation of ionization cross section for Gold L-3 as a function of energy. The numbers next to symbols correspond to references in the database bibliography.

would help encourage other workers to try and fill them.

Until more data become available, it is also necessary to decide how best to make use of the compilations that are presented here. The wide discrepancies often encountered between nominally equivalent measurements make determining a best, or even a most probable, value for the parameter of interest a difficult task. One possible solution might be to use theoretical yield versus energy curves, determined from Monte Carlo simulations or analytical fits (e.g., reference 1 in the database bibliography), as templates for a fit to the data, using an iterative approach to get the best overall correlation. Initial attempts at doing this with BSE yields, employing the empirical equations suggested by Hunger and Küchler (1979), show that this approach can be a useful way to impose some order on the results, but the fitting procedure is lengthy.

Copies of the database as a collection of ASCII files (MS-DOS format), or CRICKETGRAPH files (Macintosh) are available on request from the author.

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