

PHYSICAL REVIEW LETTERS

VOLUME 29

18 SEPTEMBER 1972

NUMBER 12

K-Shell Ionization Cross Sections for 2.04-MeV Electrons

W. Scholz and Angela Li-Scholz

Department of Physics, State University of New York at Albany, Albany, New York 12222

and

R. Collé* and I. L. Preiss

Department of Chemistry, Rensselaer Polytechnic Institute, Troy, New York 12181

(Received 5 July 1972)

K-shell ionization cross sections have been measured at 2.04-MeV incident electron energy for 32 elements from V ($Z=23$) to Bi ($Z=83$). Measured cross sections drop from a value of 353 b at V to 9.9 b at Bi. The general trend of the Z dependence of the cross sections is in agreement with theoretical predictions of Kolbenstvedt. However, variations in cross sections by as much as 30% from one element to the next are not accounted for by the theory.

Atomic ionization by electron collision has been treated extensively in the literature.¹ However, although impact ionization by low-energy electrons is amply documented,² information is sparse on electrons above 0.5 MeV incident energy. In the MeV range, *K*-shell ionization cross sections have been measured for only seven elements, mostly at bombarding energies at or below 2 MeV.³⁻⁵ For Pd and Au, data also exist for 2.5- and 7.5-MeV electrons.⁶ In the very-high-energy range of 150 to 900 MeV, cross sections have been measured for eight elements from Cu to Bi.⁷ The energy dependence of the ionization cross section was investigated in most of these experiments and found to show a rather smooth variation. The experimental results generally agree with theoretical expectations to better than 25%.⁸ In view of such overall agreement, it seems unlikely that further studies on isolated elements will add significantly to the present picture. The more profitable approach might be a systematic mapping out of the cross sections as a function of atomic number and of energy, in order that detailed comparisons with theory might be made.

In this Letter, we will present *K*-shell ionization cross sections for 2.04-MeV electrons for 32 elements from V ($Z=23$) to Bi ($Z=83$). As will be explained later on in the text, the cross sections were first determined relative to each other, and then were normalized to 43 b at Sn. By making relative measurements, systematic errors were minimized so that minor variations did not become obscured by the larger uncertainties usually associated with absolute measurements. The full details of the experiment will be published elsewhere.⁹

The 2.04-MeV electron beam was produced with the Dynamitron accelerator at the State University of New York at Albany. The beam was delivered to the target in a 10-cm-diam beam transport system. After traversing the target the electrons entered a 250-cm-long section of 10-cm-diam beam pipe which served as a beam dump. The measurement of relative *K*-shell ionization cross sections was based on the determination of relative $K\alpha$ x-ray yields. Targets were fabricated by immersing lens tissue paper (~ 1.35 mg/cm²) into solutions containing a well-determined ratio of atoms of element Z to Cd.

After the sample was dried, the amount of material adsorbed was found to be typically 1 mg/cm². As the target was irradiated the fluorescence x-rays were detected in lithium-drifted Si or Ge semiconductor counters positioned at 90° with respect to the incident electron beam. No corrections were made for the angular dependence of the emitted x rays, which has been found to be isotropic.^{4,7} To deduce the $K\alpha$ x-ray yield of element Z relative to Cd from the observed spectrum, it is sufficient to know only the Z/Cd atom ratio and the relative efficiency of the detector at the appropriate $K\alpha$ x-ray energies. It is not necessary to know the electron beam intensity, the absolute number of target atoms, the precise solid angle, or the absolute detector efficiency. The choice of Cd as the reference element was somewhat arbitrary. It was considered a good choice because its $K\alpha$ x ray of 23 keV is high enough not to be easily absorbed, but low enough still to be easily detected. In addition, it is readily available, inexpensive, and has convenient chemical properties.

The Si(Li) detector, which had a 0.025-mm-thick Be window, was vacuum coupled to the target chamber. The Ge(Li) detector had a 0.13-mm Be window, as well as a 40- $\mu\text{g}/\text{cm}^2$ Au layer on its front face. To reach the latter detector, x rays from the target had to pass through a 0.051-mm Mylar exit window and a 38.6-cm air path. A sweeping magnet was used with both systems to prevent scattered electrons from entering the detector. Efficiency calibrations for these detectors will be presented in a later paper.⁹ It might be mentioned, however, that both detectors were found to have active regions which were thinner than specified by the manufacturers.

The measured $K\alpha$ x-ray intensities were corrected for absorption in the windows and air path between target and detector. Correction factors for self-absorption in the target were determined in a separate series of off-machine measurements in the following manner. X rays were produced by fluorescing a thin target of the desired element Z . The intensity of x rays at the detector was measured with and without an interposed lens-tissue Z/Cd target, thereby determining an effective attenuation factor $\exp(-\mu t)$. Having obtained μt , the fractional target transmission T , defined as the ratio of the intensities of x rays emerging from the target to the total x rays produced, was then calculated for the pertinent geometry, namely, that of a target placed 45° to both the electron beam and the detector.

Values of T as a function of Z were found to increase smoothly from 82% at V to 98% at Sr. Errors in T were estimated to be less than 2% in all cases. From the corrected relative $K\alpha$ x-ray intensities, the K -shell ionization cross sections σ_K were derived according to the following relationship:

$$\sigma_K = N \frac{I[1 + P(K\beta)/P(K\alpha)]}{\epsilon \omega_K} \quad (1)$$

In this equation, the quantities I and ϵ are, respectively, the relative $K\alpha$ x-ray intensity and the relative detector efficiency at the $K\alpha$ x-ray energy. N is the proportionality constant which normalizes our relative measurements to $\sigma_K = 43$ b at Sn ($Z = 50$). The latter value was derived from the experimental result of 44 ± 4 b, measured at 2.00 MeV incident electron energy, by replacing the fluorescence-yield value of 0.84 used in Ref. 5 with $\omega_K = 0.859$ from Bambynek *et al.*¹⁰ All other values of the K -shell fluorescence yield needed for Eq. (1) were also taken from Ref. 10, using what are referred to as "fitted values." The uncertainty in ω_K , given as "total uncertainty $\Delta\omega_K$ " in the same reference, is propagated as an error in the present analysis. Values of the radiative transition probability ratios $P(K\beta)/P(K\alpha)$ were from Nelson, Saunders, and Salem.¹¹ Errors in σ_K arising from uncertainties in this ratio amounted to less than 1%. In this procedure, it is implicitly assumed that double inner-shell ionization is negligible. This assumption appears to be justified for the range of elements studied.^{1,12}

Results for the K -shell ionization cross section are shown in Fig. 1. The open and closed circles represent, respectively, data obtained with the Si(Li) and the Ge(Li) detector system. Where the overlap in the two sets of data is better than 2%, the points are shown as half-filled circles. The error bars on the data points include uncertainties in ω_K , in counting statistics, background subtraction, and absorption of the x rays either in the target or in other materials. The absence of an error bar implies that the uncertainty is less than 5%. With the exception of region IV ($Z > 66$), however, the error bars do not include any contribution from the efficiency calibration of the two detectors. The approximate efficiency calibration uncertainties appropriate to the regions I and III are depicted in Fig. 1 by the error bars on the points enclosed in parentheses, those for region II being too small to show on this scale. Since the errors in the efficiency calibration are

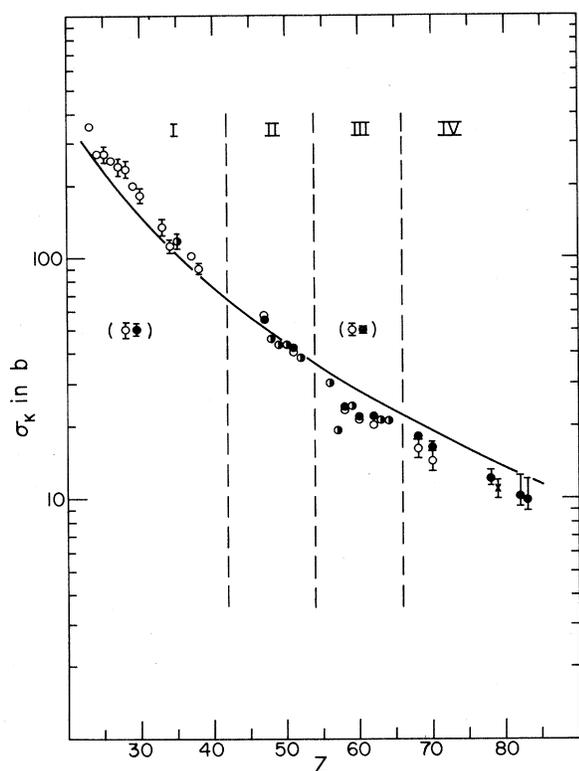


FIG. 1. K -shell ionization cross sections at 2.04 MeV incident electron energy. The data are normalized to 43 b for Sn. The open and closed circles correspond, respectively, to measurements with the Si(Li) and the Ge(Li) detector system. The half-filled circles represent data points where the overlap is better than 2%. The value for Au indicated by a cross is taken from a measurement at 2.00 MeV (Ref. 5). For a discussion of the error bars and the normalization, see text.

primarily due to uncertainties in the detector thicknesses, these errors are correlated such that the upper error limit in region I corresponds to the lower limit in regions III and IV and vice versa. Also not shown is a systematic error of $\pm 9\%$ on all cross sections resulting from the uncertainty of ± 4 b in the absolute K -shell ionization cross section for Sn as given in Ref. 5. The reason for this manner of data presentation is the following. Errors in the ratios of relative detection efficiencies increase with increasing separation of Z values. Conversely, the efficiencies for adjacent elements have very small errors relative to each other. The four regions in Fig. 1 roughly represent zones within which the data points have very small relative errors arising from detection efficiency uncertainties.

The curve drawn in Fig. 1 has been calculated using an expression given by Kolbenstvedt.⁸ The

trend of the experimental data follows the theoretical prediction rather well, especially if allowance is made for possible errors due to the efficiency calibration. The points also suitably bracket the result for Au, which is taken from Ref. 5 and indicated by a cross in Fig. 1. However, the structure in the K -shell ionization cross section around $Z=48$ and $Z=57$ cannot be accounted for by the theory which predicts a smooth behavior with Z . The observation of a pronounced dip at La ($Z=57$) is especially surprising since the following elements are also rare earths with similar physical and chemical properties. In view of these unexpectedly large fluctuations, the question of whether variations in the precise composition of the targets affect the outcome of the measurements was investigated. Several targets each of Pr, Nd, Eu, and Gd were made in which the atom ratios to Cd were varied up to a factor of 2. In addition, hydrochloric acid, nitric acid, and aqua regia were variously used for dissolving the chemicals. We found that in all instances, the data reproduced for each element to better than 2%. The effect of bremsstrahlung was also considered. By a simple order-of-magnitude calculation, it can be shown that the ratio of bremsstrahlung produced to 2-MeV electron-produced K -shell ionization is never in excess of 1.5% for any target used in the present experiment. Moreover, any effect from bremsstrahlung would again vary only slightly for adjacent elements.

In summary, the present measurements of K -shell ionization cross sections display reasonable general agreement with the predictions of Kolbenstvedt.⁸ However, sizable local variations are seen which are not ascribable to experimental uncertainties. Moreover, the most pronounced irregularities are not correlated in any obvious way with atomic shell structures. The situation seems to suggest the need for further refinements in the theoretical treatments.

We wish to thank the staff of the Nuclear Accelerator Laboratory of the State University of New York at Albany for their assistance in carrying out this work. We are also grateful to Professor Bernd Crasemann for sending preprints of work on fluorescence yields.

*Present address: Brookhaven National Laboratory, Upton, New York 11973.

¹H. S. W. Massey and E. H. S. Burhop, *Electronic and Ionic Impact Phenomena* (Oxford Univ. Press, Oxford, 1969), Vol. I.

²See, for example, B. L. Moiseiwitsch and S. J. Smith, *Rev. Mod. Phys.* **40**, 238 (1968), and references quoted therein.

³H. Hansen, H. Weigmann, and A. Flammersfeld, *Nucl. Phys.* **58**, 241 (1964).

⁴J. W. Motz and R. C. Placious, *Phys. Rev.* **136**, A662 (1964).

⁵D. H. Rester and W. E. Dance, *Phys. Rev.* **152**, 1 (1966).

⁶K. H. Berkner, S. N. Kaplan, and R. V. Pyle, *Bull. Amer. Phys. Soc.* **15**, 786 (1970).

⁷L. M. Middleman, R. L. Ford, and R. Hofstadter, *Phys. Rev. A* **2**, 1429 (1970).

⁸H. Kolbenstvedt, *J. Appl. Phys.* **38**, 4785 (1967).

This paper also contains a comparison of earlier theoretical predictions with experimental results.

⁹A. Li-Scholz, W. Scholz, R. Collé, and I. L. Preiss, to be published.

¹⁰W. Bambynek, B. Crasemann, R. W. Fink, H. U. Freund, H. Mark, C. D. Swift, R. E. Price, and P. Venugopala Rao, "X-Ray Fluorescence Yields and Coster-Kronig Transition Probabilities" (to be published).

¹¹G. C. Nelson, B. G. Saunders, and S. I. Salem, *At. Data* **1**, 377 (1970).

¹²T. A. Carlson, W. E. Moddeman, and M. O. Krause, *Phys. Rev. A* **1**, 1406 (1970).

Ultrasonic Evidence Against Multiple Energy Gaps in Superconducting Niobium

D. P. Almond, M. J. Lea, and E. R. Dobbs

Department of Physics, University of Lancaster, Lancaster, United Kingdom

(Received 22 May 1972)

The attenuation of longitudinal ultrasound along the [100] direction in Nb, resistivity ratio $R_{300}/R_0 = 5200$, has been measured with precision at low temperatures. The total attenuation in the normal state can be accurately deduced by a BCS calculation, with a single gap $2\Delta(0) = (3.52 \pm 0.02)kT_c$, from the measured superconducting attenuation. No experimental evidence has been found for the existence of either a smaller or a larger gap.

Suhl, Matthias, and Walker¹ extended the BCS theory of superconductivity to the case of two overlapping electron bands and suggested that this might apply to pure superconducting transition metals such as Nb. They postulated that each band could have distinct superconducting energy gaps and possibly different transition temperatures, depending on the strength of the interband and intraband couplings. Experimental evidence for two energy gaps in Nb came from the heat-capacity data of Shen, Senozan, and Phillips,² which were interpreted³ in terms of a *d*-band gap $2\Delta_d(0) = 3.5kT_c$ and an *s*-band gap $2\Delta_s(0) = 0.32kT_c$. The ratio of the density of states at the Fermi level, N_s/N_d , depended on the purity of the sample and was 0.015 when the residual resistivity ratio (RRR) was 110. Direct evidence for the small energy gap has also come from tunneling experiments on pure Nb crystals. Hafstrom and MacVicar⁴ deduced an energy gap $2\Delta(0) = 0.37kT_c$ from the tunneling characteristics along many directions (with the notable exception of the [100] direction). Thermal-conductivity measurements⁵ on pure Nb were initially interpreted^{5,6} on a two-gap model, but later data led Anderson, Satterthwaite, and Smith⁷ to conclude

that they provided no evidence for a second gap, and put an upper limit on $N_s/N_d \approx 10^{-3}$ for a sample with an RRR of 2000.

Recently, ultrasonic attenuation measurements^{8,9} on superconducting Nb have been analyzed in terms of a two-gap model. However, the energy gaps required to fit the data were quite different. At low temperatures the usual BCS gap $2\Delta_1(0) = 3.5kT_c$ applied, but near T_c a large gap $2\Delta_2(0) \approx 10kT_c$ was used. This large gap was purity dependent, increasing as the sample RRR increased. These two gaps were associated by Lacy and Daniel⁹ with the *s* and *d* bands, in contrast to the previous analyses,^{3,4} where the *s*-band gap was very small. Here we report measurements of the electronic attenuation in the normal state, α_n , and in the superconducting state, α_s , of a very pure single crystal of Nb at low temperatures. We show that all the electronic attenuation is due to electrons with a BCS energy gap and find no evidence for either a very small or an anomalously large one.

The normalized electronic attenuation α_s/α_n is given in BCS theory by the well-known expression $2f(\Delta)$, where f is the Fermi function and $\Delta \equiv \Delta(T)$ is the temperature-dependent energy gap.