Electrons from a 4 MeV Van de Graaff accelerator are used to produce intense sources of characteristic X-rays from a sampling of mid-to-high Z elements. Thick solid targets and high pressure gas samples are bombarded by up to 50 microamperes of 2.5 MeV electrons. The X-rays are viewed in the backward direction to reduce the background resulting from bremsstrahlung radiation. Wavelengths and widths of the X-rays are measured with a precision two-axis flat crystal spectrometer. The spectrometer is equipped with angle interferometers having an accuracy of a few tenths of a milliradian second. The lattice spacing of the crystals has been measured in terms of the wavelength of visible light by means of simultaneous X-ray and optical interferometry and is consistent with the scale used in theoretical calculations. The accuracy of the new wavelength measurements is limited to 3 to 5 ppm by the intrinsic line width of the X-rays.

Comparison of the new more reliable wavelength values with relativistic theoretical calculations has resulted in revisions of the theoretical calculations. The large discrepancies (\(\chi 10^{-1}\) eV) which existed in 1979 have been significantly reduced (\(\chi 2\) eV). Larger discrepancies for 2 > 92 suggest that additional precision measurements need to be made in this region.

Introduction

Accurate wavelengths and widths of characteristic X-rays are of interest scientifically because these measurements are used as tests of relativistic theoretical calculations and as standards in some experiments. In both of these applications it is often convenient and/or necessary to measure X-rays from elements spaced throughout the periodic table. A simplified source for producing intense characteristic X-rays is electron bombardment of a solid target containing the element whose X-rays are desired. For the measurements reported here, it is essential that the bombarding electrons have an energy of several hundred keV because high Z K series X-rays have energies around 100 keV. The efficiency of the high resolution X-ray spectrometer is very low so that electron currents on the order of a millimicroampere are needed to produce intense enough sources for reasonable data throughput. Such high currents cause appreciable heating of the targets so that provision for cooling is necessary. In addition, for a study over a wide range of Z, the targets need to be easily interchanges.

A modern target chamber equipped with a rather simple target chamber satisfies all of the above source requirements. Fortunately there are a number of electron accelerators at NBS including a 4 MeV electron Van de Graaff which had a large block of available beam time and space in the target room. A water cooled target chamber was assembled and attached to an existing beam port and a two-crystal spectrometer was moved into the target area.

**Summary**

**Accelerator**

The 4 MeV electron Van de Graaff was operated at 2.5 MeV for all of the measurements. This energy was chosen because the production of K X-rays is near maximum for bombardment of thick mid-to-high Z targets at this energy. Figure 1 shows the dependence of the K X-ray yield on the bombarding electron energy for thick targets of silver and gold. For high Z elements a significant increase in yield results from increasing the bombarding electron energy from a few hundred keV to a few MeV. Figure 1 suggests that the use of 4 MeV electrons would lead to increased yields for high Z targets. However, our measurements require that the accelerator operate in a stable manner for several hours. Stabilization of the accelerator at 2.5 MeV following conditioning of the accelerator to an energy of 3.5 to 4 MeV permitted satisfactory operation of 4 to 5 hours. Stabilization at 3 MeV reduced the operating time to about 2 hours.

The position and focus of the electron beam on the target was monitored by means of fluorescent coatings and a television camera. Beam diameters of 1 to 2 mm were typical. Because of the low efficiency of the spectrometer, background radiation levels need to be reduced as low as possible. Background radiation was reduced to a tolerable level by properly conditioning the accelerator to prevent any voltage breakdown and by carefully adjusting the beam optics to prevent the beam from striking any apertures or pipes. In addition, rather massive lead and concrete shielding is used around the target chamber, the spectrometer, and the detector. Typical background counting rates are 0.5 to 1 count/sec.

**Experiment**

![Figure 1. Dependence of the K X-ray yield on the initial electron kinetic energy for thick targets of silver and gold.](image)

The target angle, \(\alpha\), and the X-ray angle, \(\theta\), are explained in Figure 2. From Ref. 1.

U.S. Government work not protected by U.S. copyright.
The geometry of the electron beam, target, and X-ray spectrometer entrance angle is shown in Fig. 2. The target angle, α, was 30 deg and the spectrometer entrance angle, θ, was 120 deg from the beam direction. This backwards X-ray direction was chosen in order to reduce the bremsstrahlung background which is strongly peaked in the forward direction.

Targets

The solid targets were disks of 1.7 cm diameter and 0.5 to 1 mm thickness. The dependence of the K X-ray yield as a function of target thickness for gold is shown in Fig. 3. The thickness of the targets was chosen to be on the order 0.5 of the electron range which is sufficient to give maximum photon yield at 2.5 MeV. The targets were mechanically held to a water cooled aluminum target post which also served as a Faraday cup. The gas target was a high pressure (2 x 10^6 Pa) thin window cell which was also water cooled.

The target and target chamber are electrically insulated from each other and the accelerator vacuum pipes. The target and chamber currents were measured by means of an electrometer and were used to normalize the X-ray intensity. Beam currents of 20 to 50 microamperes were typical. For some targets (i.e., Pb), temperatures near the melting point were achieved in the area of the electron beam spot.

**X-ray spectrometer**

The X-ray spectrometer is a two-axis flat crystal transmission spectrometer which measures the small Bragg angles through which the X-rays are diffracted. The spectrometer is shown schematically in Fig. 2. The axes of the spectrometer are equipped with angle measuring interferometers of the Michelson type modified to be polarization sensitive. They have an accuracy of a few tenths of a milliarc second. The spectrometer operates under complete control of a computer and has an angular range of 15 degrees. The axes are calibrated in degrees by using a 24-sided optical polygon. All 24 external polygon angles are measured in terms of interferometer fringes and the sum of the angles is required to equal 360 degrees.

The crystals are nearly perfect Si wafers cut so that reflection occurs from the 220 planes. The crystals are 5 cm by 2.5 cm, but only a small central area is used for the X-ray measurements. The lattice spacing of the crystals has been measured in terms of optical wavelengths using the techniques of simultaneous X-ray and optical interferometry and lattice spacing intercomparisons of nearly identical crystals. These techniques have been described in detail in a recent review article along with the angle measuring spectrometer. The accuracy of the lattice spacing determination is 0.1 to 0.2 ppm.

X-ray profiles or rocking curves are recorded in a point-by-point manner using a NaI(Tl) detector. Profiles are recorded for the parallel (non-dispersive) and anti-parallel (dispersive) positions. The efficiency of such a two-axis flat crystal spectrometer is a few times 10^{-11} so that only very intense X-ray lines can be measured with a reasonable effort. For this reason the X-ray measurements were restricted to the Kα1, Kα2, Kβ1, and Kβ2 transitions.

**Data analysis**

The recorded profiles were fitted to model functions using a non-linear least squares computer program. The models used were a lorentzian function and a convolution of a lorentzian function with a vertical divergence function. The positions and widths returned by the fitting program were used to determine Bragg angles and linewidths. By combining lattice spacing measurements with the measured Bragg angles, X-ray wavelengths are determined.

---

Figure 2. Schematic of the electron beam, target, and X-ray emission geometry and the X-ray spectrometer.

The symbols are α - target inclination angle, θ - X-ray emission angle, S - Soller collimators, C - crystals, and D - detector.

Figure 3. Dependence of K X-ray yield ratio \( \frac{Y(t)}{Y(t=R_0)} \) on the target thickness, \( t \), of a gold target for 3 MeV electrons. \( R_0 \) equals the electron range which for 3 MeV electrons on gold is 1.2 mm. From Ref. 1.
Theoretical Comparison

Our interest in precision X-ray measurements over a wide range of Z began in 1979 and resulted from two circumstances. The first was the development at NBS of fast computers and techniques for measuring optically referenced γ-ray and X-ray wavelengths which have the capability of about 0.1 ppm accuracy. Although the new X-ray wavelength measurements have an accuracy of only 3 to 5 ppm because of large intrinsic line widths, they are in many cases a significant improvement over previous measurements. The second was the significant improvement in theoretical calculations in the last several years. These calculations are made in the relativistic self-consistent field approximation and energies of neutral atoms and ions are determined separately in order to account for the effect of the ionization on the remaining electron orbitals.

Although term values are calculated by theorists, experimentalists can only determine term values with appreciable difficulty. Generally several measured transition energies need to be combined with an outer level ionization energy so that there are accumulation of errors and difficulty in interpretation of the ionization limit. On the other hand, the theoretical calculations have achieved a high enough precision so that the subtraction of term values leads to sufficiently accurate transition energies that experiment-theory comparisons are meaningful. Thus, we have preferred to make comparisons between observed transition energies and calculated term value differences.

The most informative comparison between experimental and theoretical X-ray energies which was available in 1979 was a highly selected set of X-ray emission lines and the full Z calculation of Huang, et al. The X-ray data set included a few X-ray lines (Cu Kα1, Mo Kα1,6,7 and W Kα1) which were directly measured on the optically based scale using the techniques described here. Precision X-ray measurements using a curved crystal spectrometer calibrated with theoretically based γ-ray standards also contributed a significant number of lines to the data set in the high Z region (Kα1,2 transitions in Tm, Th, U, and Pu). Finally, wavelength ratios obtained by J.A. Bearden12 and his collaborators in the 1960's were used to further expand the data set (Kα1,2 transitions of Al, Cr, Cu, Mo, Ru, Ag, Sb, and W). A list of energy values and uncertainties and the details of the selection process are presented in Ref.4.

A graphical representation of the experimental-theoretical comparison which was available in 1979 is shown in Fig. 4. The differences between experimental and theoretical transition energies for Kα1 and Kα2 are plotted as a function of Z. The most striking feature of Fig. 4 is the nearly linear dependence of the discrepancy as a function of Z. Several of the points in Fig. 4 depart from the linear dependence by a significantly larger amount than the indicated error estimates. This fact and the limited number of data points suggested that the apparent linear discrepancy should be viewed with caution.

The comparisons presented in Fig. 4 encouraged both theorists and experimentalists to consider further precision X-ray wavelengths. On the theoretical side, Chen, et al13 revised the Huang, et al calculations by using a Fermi distribution rather than a uniform distribution for the nuclear charge, a more accurate expression for the Breit interaction which better takes into account that electron velocities may be large and a more complete self energy estimation. Although the published revision is limited to the K and L levels for Z ≥ 70 further calculations have been communicated privately so that revised theoretical estimates for all Kα1,2 transitions are available. More details concerning the revised theoretical values are available in Ref.14.

On the experimental side, a program of precision X-ray measurements was begun at NBS. The new measurements included Kα1,2 and Kβ1,2 transitions for Xe, Ba, Er, Au, Pb, Th, and U and Kα1,2 transitions for Ag, Nd, and Sm. A complete listing of the measured wavelengths, energies, widths, and uncertainties is given in Ref.14. The uncertainty of the Kα transitions is 3 to 5 ppm and of the Kβ transitions is 4 to 7 ppm. These new optically based wavelengths were again combined with other precision wavelength measurements in order that an experiment-theory comparison could be made over as large a range of Z as possible. All of the data used in the 1979 comparison described above were included except those which were remeasured in this study. In addition, curves in crystal measurements from Barreau, et al, on Cr, Mo, Kα1,2, Cu Kα1,2, cm Kβ1,2, and Ba Kα1,215 significantly expanded the data set in the high Z region. In the expansion of the data set only Kα1 and Kα1,2 were directly available for elements in order to get needed values for Kα2 and Kβ1,2, the ratios Kα1/Kα2, Kα1/Kβ1, and Kα2/Kβ1 from Ref.10 were used. A table containing energies and uncertainties of the expanded data set is available in Ref.14.

The comparison of these more recent experimental results for Kα1 and Kα2 with the revised theoretical calculations is shown graphically in Fig. 5. The most striking feature of Fig. 5 is the absence of the dominant Z dependent trend which was evident in Fig. 4. The Kα lines show a more or less constant difference of 1 to 2 eV up from Z 90. The Kα1,2 difference near zero around Z = 90 to 92 and larger (5 eV) differences around Z = 97. A possible explanation of the relatively constant 1-2 eV discrepancy is uncalculated electron correlation effects.14 For the inner K and L levels which are involved in these transitions, ground-state and hole-state configuration interaction with nearby bound states and with radiation and radiationless continua may have to be considered.
Energy differences between the most recent experimental values and the theoretical estimates of Chen, et al.\(^\text{11}\) for KaI (a) and KaII (b) emission lines as a function of Z. The symbols are: Ref. 10-12 - ; Ref. 14 - ; Ref. 8 - ; Ref. 15 - .

The experimental data presented in Fig. 5 were obtained predominantly from solid samples while the theoretical calculations are for free atoms. Comparison of experimental data which may suffer from chemical and/or solid state perturbations with free-atom calculations needs to be justified. Measurements on gaseous Xe were made to set some limits on the chemical and solid state shifts. In Fig. 5, the gaseous Xe points are in no way distinguished which suggests that the inner levels involved experience no or nearly equal shifts due to chemical and solid state effects.

In Ref. 14, plots similar to Fig. 5 but for KBI and KBII transitions are presented. Those plots which are somewhat less informative because the theoretical calculations of Huang, et al.\(^\text{5}\) have not been revised for the MII,III levels show larger discrepancies than the KI line. Preliminary calculations\(^\text{17}\) appear to suggest that the MII,III levels shift by a larger amount than the LII,III levels due to coupling to bound states and continua. A more thorough discussion of the KBI,II transitions is available in Ref. 14.

In addition to advances in the theoretical calculation of energies, improvements have been made in calculations of atomic transition probabilities and level widths.\(^\text{18,19}\) The calculations are relativistic in nature and include all possible Auger transitions. In Fig. 6 the calculated KaI line widths are compared to the line widths measured in this study. A similar plot for the KaII lines is nearly identical. The excellent agreement between theory and experiment suggests that the theoretical interpretation is quite accurate and that none of the measured emission lines is significantly broadened by satellite lines.

Figure 5. Energy differences between the most recent experimental values and the theoretical estimates of Chen, et al.\(^\text{11}\) for KaI (a) and KaII (b) emission lines as a function of Z. The symbols are: Ref. 10-12 - ; Ref. 14 - ; Ref. 8 - ; Ref. 15 - .

Figure 6. Transition energy widths. The solid line is the theoretically calculated widths (Refs. 18 and 19) and the dots are the experimentally measured widths.

Acknowledgments

We acknowledge and thank C.E. Dick and J. Mills of the Center for Radiation Research for assistance in the maintenance and operation of the Van de Graaff and B. Crasemann and M.H. Chen for providing preliminary calculations and many helpful discussions.

References


2. The electron range is defined as the average path length which the electron travels in a semi-infinite slab of target material in the course of slowing down from its initial kinetic energy to zero kinetic energy. Tables of electron ranges for different initial electron energies and materials are given by M.J. Berger and S.M. Seltzer, NASA Report No. SP-3012 (unpublished).


