Intensities of Monochromatic Continuous X-Rays from Atomic Targets of Nickel

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Intensities of continuous x-radiations from a thin target (199A) of metallic nickel under electron bombardment in the range 12 to 180 ky have been observed. Ross filters of Ag-Pd (485-508 x.u.) and Se-As (979-1044 x.u.) provided monochromatization, and measurements were performed with an argon-filled ion chamber and a quadrant electrometer. Sensitivities of the measuring system at various wave-lengths were determined and corrections were made for all absorptions in the path of the radiation. Small corrections for radiation from sources other than the target and for the several effects of the finite target thickness were applied. The corrected data show relative intensities I per unit wave-length interval for the selected wave-lengths and direction of observation as produced by electron bombardment of independent nickel atoms. The observed variation of I with bombardment energy is in excellent agreement with the Sommerfeld theory and the variation of I with wave-length is in approximate agreement.

INTRODUCTION

S a field for quantitative measurement the continuous x-ray spectrum has been long neglected. Well known for several decades have been the qualitative aspects of the spectrum which proceeds from thick targets, including the wave-length-intensity distribution and its dependence upon bombardment energy, target atomic number, and (less certainly) direction of emission. We have also possessed one quantitative fact: the law of Duane and Hunt. Although more refined and quantitative thick-target data might have a certain technical interest, pure physicists have not been attracted to this field of measurement because the thick target is so special and complex a source as to defy exact theoretical analysis. Lacking the possibility of comparing measurements with theory, observers have regarded the field as unremunerative.

The continuous x-radiation from thin, or virtually monatomic, targets under electron bombardment is susceptible of theoretical treatment,¹ being, indeed, the result of one of the most fundamental of possible collisions. There is no doubt here as to the utility of quantitative information but experimental difficulties have stood in the way, and we are at this writing very far from the ability to describe the radiation from such sources in quantitative terms. Kulenkampff,² Nicholas,³ and others have observed continuous spectra from targets less thick than the productive layer of thick targets but not sufficiently thin to classify as atomic targets. These observations have been valuable in showing the conspicuous differences between thickand thin-target spectra and in indicating qualitative agreement with thin-target theory. In none of these cases was an attempt made to deduce the precise shape of an atomic-target spectrum, nor would this have been easy, with the available data. To reduce thin-target spectra to atomictarget spectra, in the present state of our knowledge of the behavior of fast electrons in solids, it is necessary to have the target thin enough or the electrons fast enough so that the velocities of the latter (both speeds and directions) are modified by the target atoms only to that small extent for which theoretical corrections may be applied.

To meet the conditions of theory the experimenter must produce sufficiently thin targets and he must use observing means sensitive enough to measure the feeble radiation after spectral resolution. Targets of evaporated and condensed nickel satisfied the first condition in the present work. Because of their great advantage in light-gathering power we have used Ross⁴ filters for isolating the radiation bands to

¹ A. Sommerfeld, Ann. d. Physik 11, 257 (1931); F. Sauter, Ann. d. Physik 18, 486 (1933); G. Elwert, Ann. d. Physik 34, 178 (1939); R. Weinstock, Phys. Rev. 61, 584 (1942).

² H. Kulenkampff, Ann. d. Physik 87, 579 (1928).

³ W. W. Nicholas, Bur. Stand. J. Research 2, 837 (1929). ⁴ P. A. Ross, Phys. Rev. 28, 425 (1926); J. Opt. Soc. Am. and Rev. Sci. Inst. 16, 433 (1928).

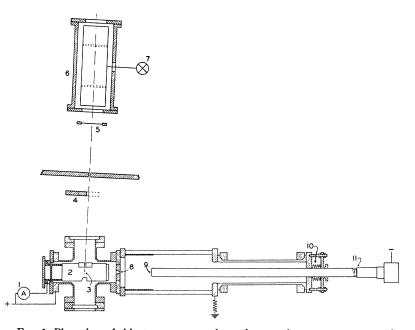


FIG. 1. Plan view of thin-target x-ray tube and measuring apparatus approximately to scale. 1. Microammeter reading current to target. 2. Hollow aluminum cylinder containing target. 3. Nickel-on-cellulose acetate target. 4. Clock-controlled shutter. 5. Balanced filter and iron prefilter. 6. Argon-filled ion chamber. 7. Electrometer. 8. Anode plate, perforated to pass electrons to field-free region containing the target. 9. Cathode filament. 10. Sylphon to permit aiming of electron stream. 11. Cathode focusing adjustment controlling shield at 9. Overall length of x-ray tube = 130 cm.

be studied. With these filters the radiation was intense enough for measurement by standardized ionization methods. Nickel was selected as the first target element in a projected series of studies covering a wide range of atomic numbers. This program as a whole would include intensity and polarization measurements with a variety of atomic numbers, electron energies, wave-lengths, and angles of observation.

APPARATUS

The nickel target was produced by a modification of the evaporation technique described by Pockman and Webster.⁵ A backing film of cellulose acetate 940A in thickness was mounted in vacuum 10 cm above a nickel ribbon which could be heated, to its melting point if desired, by a well-controlled alternating current of about 100 amperes. It was possible to keep track of the progress of the deposit by measuring through photoelectric means the progressive darkening of a glass window of the evaporation chamber. By previous calibration the constant relation between the window opacity and the surface density of the metallic deposit on the acetate film was known. All data to follow were obtained from a target whose calculated thickness was 199A.

During the evaporation the target was supported on a light and flexible structure of aluminum wires as described by Pockman and Webster.⁵ The completed target on its supports was transferred for use to the x-ray tube shown in Fig. 1 where it is seen to be mounted across the axis of a deep aluminum cup. The cup served to catch that great majority of the bombarding electrons which the target failed to stop; it was made of a substance of low atomic number to facilitate retention of the electrons and it was insulated, except for the microammeter lead, because it was essential to measure the current *to or through* the target.

It was an important feature of the design in Fig. 1 that the fragile target was inclosed in a

⁶L. T. Pockman and D. L. Webster, Rev. Sci. Inst. 12, 389 (1941).

field-free metal box, protecting it at all times from the kind of destructive gas bursts which were the ruin of so many targets during earlier researches in this laboratory. One wall of this protective enclosure was penetrated by a 4.8-mm hole through which electrons of the desired energy were injected, to fall almost normally upon the target surface. The cathode which opposed this hole at a distance of about 10 cm (subject to adjustment) was equipped with a precisely controllable focusing shield and aiming mechanism which made it possible to direct a large fraction of the total emission through the hole. Those electrons which struck metal around the hole developed heat (a maximum of 50 watts) which was removed by a current of oil flowing under pressure within the hollow wall.

Two pairs of Ross filters were used and data were thereby obtained concerning the intensities of two spectral bands, a wide variety of electron energies being used in each case. Filter elements and pass band wave-lengths were Ag-Pd (485-508 x.u.), and Se-As (979-1044 x.u.) As the preparation of these and other filters is to be described in a separate publication, little need be recorded here except that the Ag-Pd filters were metal sheets and the Se-As pair were thin plates of the powdered metals in paraffin, all filters being practically of optimum thickness. The two members of each pair were balanced for equal transmission of radiation outside the pass band by rolling or pressing. The precision of balance was such that the difference between the intensities transmitted by two balanced filters at selected wave-lengths lying without the pass band was about $\frac{1}{2}$ percent of the incident intensity at those wave-lengths.

In most applications of balanced filters the effective thickness of a filter may be varied by tilting it so that the radiation passes through obliquely. In our case the size of the focal spot was such that the beam incident upon the filters was appreciably non-parallel. To tilt a filter transmitting such a beam is to vary the thickness in different ratios for the different incident rays, an operation almost completely incompatible with accurate balancing. For this reason the filters used in this work were brought to balance with their planes parallel and were always used in this way.

In using these filters an iron prefilter of known transmission was placed in the beam to minimize any effect caused by possible departure from perfect balance in the spectral region of the nickel K lines.

The ionization chamber was designed to permit comparable intensity measurements at different wave-lengths. Specifically, the collectors were flat and parallel aluminum plates separated sufficiently to prevent interference with photoelectrons, and provided with guard plates which insured precision in the location of the limits of the collecting region, 15.2 cm in length. Large Cellophane windows at both ends facilitated geometrical alignment, while contributing entirely negligible amounts of secondary radiation to the collecting region, 8.61 cm distant. The chamber was filled with dried tank argon to a pressure 18 mm below the existing atmospheric pressure, and mounted with its entrance window 98 cm from the center of the target focal spot. The direction from target to ion chamber made an angle of 93.5° with the direction of bombardment. Ionization charges were measured with a quadrant electrometer operating at a moderate and continually checked charge sensitivity.

The filters were mounted just before the entrance window of the chamber where a hole in a lead plate defined the beam area in such a way that the calibrated regions of the filters were effective and that the beam traversing the chamber lay midway between the plates. A careful survey along the beam assured that the entire focal spot could irradiate the chamber window and that a minimum of other matter could contribute either primary or secondary *x*-radiation. A test of this latter condition will be described below.

ADJUSTMENTS AND MEASUREMENTS

By varying the adjustment of the focusing shield of the cathode and applying a small bias voltage between the shield and the filament the diameter of the focal spot was kept at 1 cm for all tube potentials used in this research, a range from 8 to 180 kv. This constancy of focal spot size was perhaps not essential but it avoided the extremes of a spot so large as to include the

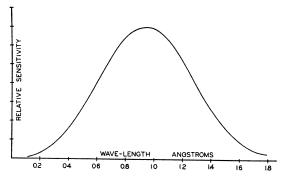


FIG. 2. Sensitivity curve of measuring system. Ordinates are proportional to collectible ionization charges resulting from emission toward the chamber of equal quantities of radiation energy.

target supports or other thick material and one so small as to produce, at the allowable current density, insufficient radiation for good measurements. As it was, the target was bombarded with a current of some 200 microamperes for the hundred hours or more required by these measurements without any observable reduction of the ample radiation output. The radiation measured in this investigation proceeded from the target in a slightly diverging beam whose axis made an angle of 93.5° with the forward direction of motion of the bombarding electrons.

In a typical run for the collection of intensity data the tube was operated on the pumps until a pressure of at most 10⁻⁶ mm Hg could be maintained with an applied voltage of 180 kv and the normal target current flowing. At voltages above 80 kv a 5-megohm resistor was connected in series with the tube at the anode end to reduce the violence of the occasional gas burst. The precise value of this resistor was checked at the beginning and end of every run and its voltage drop deducted from readings of the high potential voltmeter to obtain the actual bombardment voltage. While one operator devoted his attention exclusively to precise maintenance of the selected tube voltage and current another observed the target current and operated the filters and electrometer, recording the ionization charges accumulated in clock-controlled exposure intervals with one and then the other of two balanced filters in the beam.

Readings were not affected by electrometer drift since the electrometer was kept grounded and detached from the ion chamber until the exposure was concluded, at which time the chamber collector was connected to the electrometer, producing a ballistic deflection. As only a fraction of the tube current passed by way of the target, a special microammeter was mounted in a shielded case at the potential of the anode compartment so as to carry only the target current, and the mean reading of this instrument for every exposure was recorded.

After five or six readings with each filter the tube voltage was lowered and observations repeated at the new setting, continuing until the excitation voltage of the filter band was reached. There resulted an isochromat curve for the filter band wave-length, but a curve in need of the corrective treatment described below in order to bring out its fundamental significance. The curves to follow are the combined result of some fifteen runs of this kind.

INTERPRETATION OF DATA

Though no intensity measurements in absolute units were attempted it was desired to deduce from the data the isochromats of the free atom with all ordinates on the same arbitrary intensity scale. This required correcting the data for the unequal response of the ion chamber to different wave-lengths, for the presence of radiation from sources other than the target, and for the several effects of the finite bulk of the thin target upon speed and direction of the bombarding electrons, effects themselves speed-dependent.

In an ion chamber with guard plates the radiation must undergo absorption in a preliminary gas volume before arriving at the collecting region. The preliminary absorption necessarily makes the chamber insensitive to radiations of long wave-length. On the other hand the finite length of the collecting region makes for a low sensitivity to short wave radiations. The result is a rather symmetrical sensitivity curve like Fig. 2, which represents the result of calculations on the chamber of this investigation, calculations taking account not only of the above considerations but also of the small effects of scattering and fluorescence within the chamber, and of the reduction of intensity by absorptions external to the chamber. We are involved with scattering and fluorescence effects because the fraction of the entering radiant energy which is scattered out of the collecting region and fails therefore to contribute any ions to the collected charge is a function of wave-length, and because the escaping argon K radiation energy introduces a loss dependent upon wave-length. The method of making these corrections is essentially that of Allison and Andrew⁶ with simplifications which our particular gas, wave-length range, and chamber design permitted.

The absorbers outside the chamber included the target itself, thin windows of the tube and chamber, intervening air, prefilter, and balanced filters. By taking due account of the transmission ratios of these elements we arrive at the over-all sensitivity curve of Fig. 2, any two of whose ordinates stand in the ratio of the ionization charges which would be collected at the chamber as a result of primary emission toward the chamber of equal energies of radiation having, respectively, the wave-lengths denoted by the corresponding abscissas. It is improbable that in

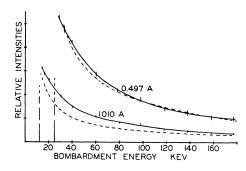


FIG. 3. Relative intensities of monochromatic x-rays from independent nickel atoms under electron bombardment. Ordinates are proportional to continuous spectrum radiant power per unit solid angle per unit wave-length interval for the selected direction of observation. Solid curves are experimental results, with probable errors of observation denoted by vertical line segments. Broken curves are theoretical (Sommerfeld-Weinstock). Theory and experiment have been brought into agreement at one point by arbitrary manipulation of the ordinate scale.

Fig. 2 the ratio of any ordinate to the maximum ordinate is in error by more than 0.01. To put observations on the same intensity scale the value of each collected charge was divided by the appropriate ordinate of Fig. 2.

Allowance for radiation from other sources than the target had to be deducted from the intensity data. The target was removed and the tube operated as before with the result that with all filters removed small but observable intensities were produced at all voltages. The intensities being insufficient for monochromatic measurements, corrections were calculated by assuming for the radiation received at each voltage a Wagner and Kulenkampff⁷ thick-target spectral distribution and normalizing the total to agree with the observations. The background intensity contributions lying within the Ross filter pass bands were then readily deduced and applied to the thin-target intensity data as subtractive corrections, the magnitudes of which ranged from 0.1 to 3 percent of the thin-target intensities.

Although our target was so thin that nearly all the bombarding electrons emerged with deviations of only a few degrees due to multiple scattering, and with energy losses of the order of one percent there were nevertheless four separable effects of target thickness which affected the continuous spectrum intensities. Of these the velocity loss has been mentioned. Its magnitudes for various bombardment energies were calculated by Williams's⁸ empirical stopping power formula, effective bombardment voltages for the target as a whole being calculated and used in plotting the final results. The other three target effects are processes which cause the bombarding electron's actual path in the target to exceed the normal target thickness. This may result from multiple scattering, from single scattering, or from return of the electrons into the target after large angle scattering by the cellulose acetate backing. The average actual path lengths in the target were calculated by a combination of the Rutherford⁹ scattering formula and Bothe's¹⁰ formula for the most probable angle of scattering. At voltages above 50 kv the calculated path lengths exceeded the target thickness by only about one percent but at 15 kv the increased multiple scattering had raised the excess to about 8 percent. The details of these calculations are elaborate and laborious and are omitted here because of the smallness of the resulting corrections and because they will be more extensively

S. K. Allison and V. J. Andrew, Phys. Rev. 38, 441 (1931).

⁷ E. Wagner and H. Kulenkampff, Physik. Zeits. 23, 503 (1922).

 ⁸ E. J. Williams, Proc. Roy. Soc. A130, 310 (1930).
⁹ E. Rutherford, Phil. Mag. 21, 669 (1911).
¹⁰ W. Bothe, Handbuch der Physik, ed. 1927, Vol. 24, 15-18.

treated in a forthcoming paper to be submitted to this journal by Webster, Pockman, Harworth, and Kirkpatrick.

After all data were obtained the target was examined and found in good condition. It was quite flat and exhibited a well-defined, darkened focal spot 1 cm in diameter. Under the microscope numerous minute holes could be seen, occupying a total of perhaps 2 percent of the focal area. Aside from this observation no attempt was made to evaluate the thickness distribution over the focal spot. It is realized that if thin spots as well as perforations existed our target corrections are somewhat too large since they were based upon an assumed thickness of 199A, the value obtained from the known loss of nickel from the filament in the evaporating process. We were watchful at all times, however, for decreases in the radiation output of the target and since none was observed, it is concluded that only negligible amounts of metal were lost during operation.

RESULTS

In Fig. 3 the corrected isochromats are plotted as solid lines with the statistical probable errors of the ordinates denoted by short vertical segments. The errors of the abscissas are negligible. These curves are in striking contrast to the nearly straight lines of positive slope which are the isochromats of a *thick* target. Recently through the work of Weinstock¹ the Sommerfeld theory of the continuous x-ray spectrum has been stated in a form which makes comparison with experiment possible with only a moderate amount of straightforward calculation and in Fig. 3 the predictions of this theory are shown as broken lines, which have been normalized by arbitrary adjustment of the ordinate scale to produce a compromise fit of theory to experiment in the case of the 0.497A curves. In comparing the shapes of the curves we consider that the agreement is all that might have been hoped for in view of the approximations of the theory (neglect of screening and relativity) and the errors of the experiment. As to the dependence of intensity upon wave-length (the vertical separation of the isochromats) theory indicates a more pronounced wave-length variation than we observed and the discrepancy here is beyond the expectations and, for the present, is left unexplained.

Seeking empirical formulas for representation of the experimental results one finds that the isochromats are approximately equilateral hyperbolas, especially the 0.497A curve, and that all of the observations may be approximated by $IV\lambda^{\frac{1}{2}}$ = constant where *I* is the observed intensity within a fixed small wave-length interval, *V* is the bombardment energy, and λ the wavelength. The inaccuracies of fit exceed the errors of observation only near the high frequency

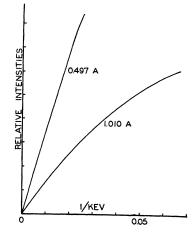


FIG. 4. Relative intensities of monochromatic x-rays from independent nickel atoms, plotted to show the observed inverse proportionality of intensity to electron bombardment energy. The validity of the approximation is evidently higher when frequency and bombardment energy are high.

limits, where the intensities fall off somewhat. This simple descriptive formula may hardly hope for theoretical justification and it is advanced only tentatively and subject to adjustment in view of future results. Figure 4 presents the experimental data of Fig. 3 in such a way as to display this empirical inverse proportionality of intensity to bombardment energy.

Naturally the *theoretical* isochromats may be represented by a similar abbreviated expression but in this case the exponent of λ lies between $\frac{3}{2}$ and 2. It is interesting that twenty-five years ago Webster¹¹ was able to deduce from thick-target data the conclusion that the intensity in the thin-target spectrum should vary inversely with λ^2 , a conclusion here approximately substantiated by both experiment and an adequate theory.

¹¹ D. L. Webster, Phys. Rev. 9, 220 (1917).