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RADIATIONS OF CERTAIN SYNCHROTRON-INDUCED RADIOACTIVITIES

By
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June 1952

Ames Laboratory
PHYSICS

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RADIATIONS OF CERTAIN SYNCHROTRON-INDUCED RADIOACTIVITIES

by

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and

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SUMMARY

This work has borne out the usefulness of scintillation counter techniques in connection with radioactivities produced by synchrotron irradiation. The activity of the sources produced has been ample for obtaining good statistical accuracy in short counting periods.

The method developed for preserving and mounting sodium iodide made it practical to take advantage of these crystals for gamma-ray energy measurements. Coincidence techniques involving gamma-rays were also improved by the use of these crystals.

The investigation of the radioactivity of Ag$^{112}$ and Ag$^{113}$ showed how the various scintillation counter techniques could be applied to a specific problem. The Ag$^{112}$ gamma-ray energy was determined using a sodium iodide crystal. The presence of beta-ray in coincidence with this gamma-ray was established and the beta-ray end point determined using coincidence methods. The Ag$^{113}$ beta-ray end point was determined with the aid of a coincidence method for reducing back-scattering. The branching ratio of the two Ag$^{112}$ beta-ray components was estimated and the corresponding $\eta$ values calculated. The values of $(W_0^2 - 1)\eta$ for Ag$^{112}$ indicate that the two beta-components may be forbidden of the type $\Delta I$ equal two with a change in parity. The Kurie plots obtained were not good enough to indicate whether this was true or not. Further analysis of the Ag$^{112}$ beta-ray components with a magnetic lens spectrometer seems desirable to furnish additional information on their forbiddeness, as well as to give better values for the branching ratio and for the end

1This report is based on a Master's thesis by Streatfield H. Cox, Jr., submitted June, 1952.

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pointso. Probably the chief contribution the scintillation counter made to this problem is the establishment of the inner beta-ray of Ag$^{112}$ in coincidence with the gamma-ray. This would be difficult to determine with the lens spectrometer.

Ag$^{112}$ was found to decay directly to the ground state of Cd$^{112}$ in 63\% per cent of the transitions with a beta-ray spectrum whose end point was 3.93\pm0.10 mev. For the other 37\% per cent of the transitions a gamma-ray of 0.62\pm0.01 mev and beta-rays with a maximum energy of 3.26\pm0.10 mev were found to be in coincidence.

A beta-ray end point of 1.86\pm0.05 mev was found for Ag$^{113}$.

### INTRODUCTION

Scintillation counters are proving to be very useful for studies in nuclear physics. There are a number of factors which contribute to their utility. The energy, location (to within the size of the crystal) and time of arrival of a given particle or photon may be determined simultaneously. Thus angular correlation of simultaneously emitted particles as well as other coincidence and energy measurements are possible. Fifty and sometimes one hundred per cent geometry can be obtained for counting a radioactive source. When combined with photographic recording techniques or used with multi-channel differential discriminators, scintillation counters enable one to determine the energies and half lives associated with the disintegration of very weak sources, including those with short half lives. The method is thus useful for the study of activities produced by betatron or synchrotron irradiations. By using anthracene or other organic phosphors, beta-ray energies can be measured with little interference from gamma-rays, while the use of absorbers and sodium iodide phosphors enable gamma-ray energies to be determined in the presence of beta-ray components. Liquid scintillators make possible the use of large quantities of scintillating material for the study of cosmic rays and other high energy phenomena.

With such varied applications numerous experimental techniques must be developed for using scintillation counters. This work concerns several of these techniques developed
for the study of the radioactive sources produced in the synchrotron. Thallium activated sodium iodide crystals are hygroscopic and require special care in handling and use, since any surface film impairs the resolution obtainable from them. A method for processing and permanently mounting these crystals has been developed. When anthracene crystals are used for beta-gamma coincidence work, the coincidence counting rate is quite low. The use of a sodium iodide crystal as the gamma detector improves the coincidence counting rate, thus making it possible to obtain enough counts to obtain good statistical accuracy in a short period of time. The distortion of beta-spectra due to backscattering of the electrons out of the crystal presents a serious handicap to the study of beta-spectra with a scintillation counter. A method for reducing this effect has been developed.

The radioactivity of Ag\textsuperscript{112} and Ag\textsuperscript{113} produced in the synchrotron provides an opportunity for the use of all these techniques and serves as an illustration showing how a scintillation counter may be used effectively to study complex decay schemes and combined beta-spectra.

The various counting techniques will be described, followed by their application to the silver problem.

The electronic equipment used in this work is essentially that developed by Palmer and described in his thesis (1).

TECHNIQUES FOR USING SODIUM IODIDE CRYSTALS

The use of thallium activated sodium iodide crystals for the determination of gamma-ray energies is well covered by the literature (2), (3). A number of various methods for surfacing and mounting these highly hygroscopic crystals has been described, most of which have one or more of the following drawbacks: they do not give good resolution, they are messy, or they are not permanent. The method to be described here is largely a combination of desirable features of other methods, but has none of the above mentioned drawbacks.

Due to their possibly poisonous nature, these crystals are handled with care, usually with rubber gloves. The breathing of any dust from the crystals is avoided. In
cases of prolonged exposure to the air the crystals are protected by immersion in mineral or silicone oil.

Preparation of the Sodium Iodide Crystal for Mounting

A rough 1 1/2 in. x 1 1/2 in. cylinder of thallium activated sodium iodide was obtained from the Harshaw Chemical Company. The crystal was removed from its shipping container and chucked in a lathe. The base of the cylinder was carefully shaped to fit the 5819 phototube. Great care was taken not to fracture the crystal. The crystal was removed from the lathe and the remaining surfaces were sanded until they were smooth although still cloudy.

The cloudy surface was next dissolved off with ethyl alcohol. Stock 95 per cent alcohol was found to be satisfactory. A number of beakers of a size sufficient to accommodate the crystal easily were filled with the alcohol. The crystal was placed in the first beaker and moved about until the alcohol became cloudy. Then the crystal was placed in the second beaker of alcohol and the process repeated and so on until the surface became completely clear. The crystal was next submerged in thin silicone oil, removed, and all remaining traces of alcohol were wiped off. Immediately the crystal was placed in silicone oil from which all air had been removed. The air had been removed by placing a beaker of the oil in a vacuum desiccator which was then evacuated with a vacuum pump.

Mounting the Crystal on the Phototube

A sleeve was first made by bending a rectangular sheet of aluminum around the phototube. This was shaped to fit the tube envelope snugly and the open end was cut to extend 1 1/2 in. beyond the sensitive end of the tube. The seam was sealed with scotch electrical tape. The sodium iodide crystal was removed from the thin oil and most of the oil was wiped off. Heavy (6830 centipoises) silicone oil was placed on the end of the crystal which was cut to fit the phototube and the crystal was placed on the tube. Magnesium oxide (light reflector) and magnesium perchlorate (dehydrating reagent) were next packed between the crystal and the aluminum sleeve. An effort was made to keep the magnesium oxide next to the crystal. The shiny side of a piece of
aluminum foil was placed on the top surface of the crystal and a cap of thin sheet aluminum placed over the end of the sleeve. All seams were sealed by wrapping with scotch electrical tape. Tube crystal assemblies made in this manner have been found to keep for months with no change in resolution. Twelve per cent resolution (full width at half maximum) was obtained for the 663 kev $^{60}$Co  gamma-ray with an average photomultiplier tube. The completed assembly is shown in Figure I.

COINCIDENCE SPECTROMETRY

Some radioactive nuclei decay emitting a beta- and a gamma-ray simultaneously. This may be determined by counting coincidences between beta- and gamma-rays. The coincidence beta-spectrum can also then be obtained without interference from other beta-ray components. The experimental setup for coincidence spectroscopy is shown in Figure II. The two crystals are placed as close together as possible to increase the coincidence counting rate. Each tube and crystal assembly is enclosed in a light tight housing. The absorber used is just thick enough to stop the highest energy beta particles from the particular source being used. The coincidence scaler reading gives the inner beta-spectrum. The beta scaler reading gives the total beta-spectrum and is used to obtain the accidental coincidences which are subtracted from the coincidence scaler reading. The differential discriminator for the gamma side is generally set to count the region of the photoelectric peak of the desired gamma-ray (provided the energy of the gamma-ray is low enough for a good photoelectric peak). This procedure tends to reduce coincidences due to extraneous effects. For example, a beta particle may radiate (bremsstrahlung) and cause a coincidence, or a gamma-ray and its compton electron may cause a coincidence.

REDUCTION OF BACKSCATTERING IN BETA-SPECTRA

Distortion of the Beta-Spectrum

The conventional setup for obtaining beta-spectra with a scintillation spectrometer has the source located on or
Fig. 1—Sodium iodide crystal mounted on the photomultiplier tube.
Fig. II--Geometry and block diagram for coincidence counting.
above a plane crystal surface. The spectrum thus obtained is found to deviate somewhat from the known spectrum. After corrections for the resolution of the instrument have been made the major part of the distortion is still found to be present. This distortion becomes progressively worse at the low energy end of the spectrum, as shown in Figure III. Although Kurie plots which give accurate end points can be made utilizing the high energy portions of the curve for simple beta-spectra, studies of the shape of spectra and of more complex spectra are in general seriously handicapped by this effect.

This distortion is thought to be caused chiefly by electrons which lose part of their energy in the crystal and then are scattered back out of the crystal. This results not only in the loss of a count at the correct energy, but also in the gain of a count at an incorrect lower energy.

Reduction of the Effect

There is some experimental evidence which at least indicates that the percentage of incident electrons backscattered by a plane surface is independent of the initial electron energy in the energy range concerned (4), (5). If this is true, it should be possible to catch the backscattered electrons in another crystal, use the resultant pulse to eliminate counts due to backscattering, and thereby obtain an improved spectrum.

The experimental setup which was used for doing this is shown in Figure IV. The "spectrum" tube, crystal, amplifiers, discriminator, etc., operated in the usual manner. The center of the coincidence crystal was hollowed out slightly to accommodate the source. The faces of the "spectrum" and "coincidence" crystals were separated only by a sheet of aluminum leaf so that no backscattered electrons could escape the apparatus. To obtain the corrected spectrum the coincidence counting rate was subtracted from the spectrum counting rate for each setting of the spectrum discriminator.

 Corrections were made for coincidence pulses below the minimum voltage which could be detected. A discriminator was placed between the amplifier and the coincidence circuit
Fig. III—Distortion of typical beta-spectrum by simple-single crystal scintillation counter.
Fig. IV--Geometry and block diagram for two crystal anti-coincidence method.
shown in Figure IV. This could be set to count all pulses above a certain minimum voltage, or else all pulses in a voltage window whose width and height could be varied. The gain of the amplifier in the coincidence side was always set as high as feasible for stable operation. The coincidence discriminator was then adjusted to count only the pulses in a window with a width of a few volts. With the setting of the window of the spectrum discriminator at a typical voltage, the height of the coincidence discriminator window was varied and the counting rates were taken at various voltages near the minimum. The curve resulting from plotting the position of the window in the coincidence discriminator against the counting rate given by the coincidence scaler was found to approach the minimum voltage as a straight line with only a slight negative slope, to a first approximation. It was thus possible to account for all beta-rays scattered into the second crystal by first making one reading counting all coincidence pulses above the minimum voltage and then by taking another reading counting all the pulses between the minimum voltage and twice the minimum voltage and adding the two together. This was done for each energy setting of the spectrum channel. The extrapolation amounted, in magnitude, to a correction of about 10 per cent to the observed coincidence rate at each setting.

Corrections were also made for accidental coincidences. This was done experimentally rather than by a calculation using the dead time. The two crystals were separated, and individual sources were provided for each. Individual counting rates comparable to those in the particular spectra under investigation were obtained by changing the respective discriminator settings. The coincidence counts under these conditions were assumed to be accidental, and were therefore subtracted from the coincidences recorded under the same conditions with actual spectra.

Application to the Beta-Ray Spectrum of $^{32}$P

To check the operation of the equipment and the effect of the method on a typical beta-spectrum, the well known simple allowed spectrum of $^{32}$P was run. The spectrum corrected for backscattering was further corrected for the resolution of the instrument as described by Palmer (1). A Kurie plot (6) was made of the result and is shown along with the corrected spectrum in Figure V. Comparison with single crystal results show that the straight portion of
Fig. V--Beta-spectrum and Kurie plot of $^{32}P$. 
the Kurie plot is lengthened and the deviation of the remainder of the points from the straight line is reduced by this method, as shown by Figure V. The 0.626 mev Cs$^{137}$ (7) conversion line was used for calibration. This gave an end point of 1.76±0.05 mev for the P$^{32}$ beta-ray.

LINEARITY OF THE INSTRUMENT

In order to check the linearity of the instrument in actual practice a composite gamma-ray source was made to provide 323±5 kev Cr$^{51}$ (8), 663±6 kev Cs$^{137}$ (7), and 1.118 mev Zn$^{65}$ (9) radiation. Thus four points (including the origin) were obtained for the linearity curve, of which a typical one is shown in Figure VI. The corresponding voltage peaks in this case were 20.2±0.3, 41.5±0.4, and 69.5±0.4 volts. Similar curves were plotted for various phototube voltages, thus varying the size of the pulses put into the amplifying equipment. For large pulses the curves were found to flatten off somewhat at high energies. All work reported in this paper was done with pulses well within the linear range of the instrument. The limits of variation of the ratio (particle energy/pulse amplitude) was found to be ±2.5 per cent.

RADIOACTIVITY OF Ag$^{112}$ AND Ag$^{113}$

The presence of a 3.2 hour activity attributable to Ag$^{112}$ and a 5.3 hour component attributable to Ag$^{113}$ was confirmed by Palmer (1) using samples prepared as described below. Palmer also obtained end points for the Ag$^{112}$ and Ag$^{113}$ beta-rays. An 860 kev gamma-ray for Ag$^{112}$ has been reported by Seiler (10). It was desired to determine the decay schemes for Ag$^{112}$ and Ag$^{113}$, and to investigate the forbiddenness of the various beta-spectra involved in this work.

All silver samples used were obtained by bombarding CdCO$_3$ with bremsstrahlung, which extends to 70 mev from the Iowa State synchrotron. The resulting silver activities
Fig. VI--Typical linearity curve for the scintillation counter.
were separated chemically from the remainder of the sample and mounted. The separation took from one to two hours, so that short-lived activities died out before the sample was placed in the scintillation counter.

Determination of the $^{112}\text{Ag}$ Gamma-Ray

The sample was bombarded for six hours and mounted on an aluminum counting planchet after the chemical separation. Three aluminum plates 1/8 in. thick were used as absorbers and placed between the source and the sodium iodide crystal. The spectrum was run using a three volt window whose position was moved in steps from the high energy end to the low and back again, while two minute counts were taken at each point. The geometric mean of the counting rates at each scale setting was plotted as shown in Figure VII. The 1.118 mev Zn$^{65}$ gamma-ray was used for calibration, which resulted in an energy of 622 kev for the $^{112}\text{Ag}$ gamma-ray. This was very close to the 663 kev Cs$^{137}$ gamma-ray energy so the two peaks were compared directly. Energies of 622 and 623 kev were obtained for the $^{112}\text{Ag}$ gamma-ray from this direct comparison with Cs$^{137}$. Comparison of counting rates taken at the same energy at different times verified that the gamma-ray was part of the 3.2 hour activity.

$^{112}\text{Ag}$ Beta-Spectra

It was next desirable to see if there were a beta-ray component in coincidence with the gamma-ray of $^{112}\text{Ag}$. The source was given a six hour bombardment and mounted in the center of a thin formvar-polystyrene film which was on a two inch diameter ring. This source was sandwiched between a 1/8 in. lead absorber and the anthracene crystal, as shown in Figure IV. The 2.23 mev (11) end point of $^{90}\text{Y}$ was used as a calibration. Corrections were made for the resolution of the instrument and for accidental coincidences. An end point of 3.26 mev was found for the inner beta-ray of $^{112}\text{Ag}$, as shown by Figure VIII.

The total beta-spectrum was run using the anthracene crystal after the absorber and sodium iodide crystal were removed. The resulting Kurie plot is shown in Figure IX. An end point of 3.93 mev for the outer beta-ray was obtained.
Fig. VII—Ag$^{112}$ gamma-ray spectrum.
Fig. VIII--Ag$^{112}$ inner beta-spectrum obtained by coincidences.
Fig. IX--Ag\textsubscript{112} total beta-spectrum with inner beta-spectrum obtained by subtraction.

\[ \left( \frac{N}{W_{\gamma}} \right)^{1/2} \]

PULSE HEIGHT - VOLTS

\[ 2.23 \text{ MEV} \]

\[ 3.33 \text{ MEV} \]

\[ 30, 32, 38, 42, 46, 50, 54, 59, 62, 66, 70, 74, 78, 82 \]
The proposed decay scheme of Ag\textsuperscript{112} is shown in Figure X. We note that the difference in energy of the two beta-rays is 670 kev rather than the 622 kev obtained by direct measurement. This represents an error of the order of one per cent for the end points of the beta-rays, so that it is not unexpected. This is especially true since the outer beta-ray Kurie plot was determined principally by three points, two of which had previously been corrected for the resolution of the instrument as described by Palmer (1). The gamma-ray energy determination was so near to that of the cesium calibration line that it should be quite accurate.

The Kurie plot of the inner beta-ray was also obtained by continuing the Kurie plot of the outer beta-ray back and making a Kurie plot of the difference in counting rates represented by this curve and the total spectrum curve. The resulting end point was 3.32 mev. Using the Kurie plots, the beta-spectra for the inner and outer beta-rays were obtained and their areas compared to obtain the branching ratio. Ag\textsuperscript{112} was found to decay directly to the ground state of Cd\textsuperscript{112} for 63 per cent of the transitions. The resulting half life for this mode of decay was 5.1 hours. The value of \( f \) (12) was \( 1.46 \times 10^8 \) seconds. \( (W_0^2 - 1)f \) was \( 1.13 \times 10^{10} \) seconds, indicating that the transition may be forbidden of the type \( \Delta I \) equal two with a change in parity (13). Thirteen per cent of the transitions go to an excited state of Cd\textsuperscript{112}. The half life of this mode of decay was calculated to be 8.6 hours, \( f \) was \( 8.15 \times 10^7 \) seconds, and \( (W_0^2 - 1)f \) was \( 0.47 \times 10^{10} \) seconds, indicating that this may be a forbidden transition of the same type (13).

The results of attempts to make spin and parity assignments and to check with the nuclear shell model were indefinite. If both beta components are forbidden of the type \( \Delta I \) equal two with a change in parity, one is led to a transition of \( \Delta I \) equal four with no change in parity for the gamma-ray. This transition would have a half life of the order of \( 10^{-3} \) seconds (14). Since the resolving time used for obtaining the coincidence beta-spectrum was about two microseconds, this type of transition is very unlikely. Further information as to the shape of the spectra from lens spectrometer data is being awaited before further work is done in this connection.
Fig. X--Proposed decay scheme for Ag$^{112}$. 
Ag\textsuperscript{113} Beta-Spectrum

Using the Two Crystal Method for Reducing Backscattering

For this determination the sample was irradiated for 11 1/2 hours and then allowed to stand for 12 hours so that the 3.2 hour Ag\textsuperscript{112} activity would die out with respect to the 5.3 hour Ag\textsuperscript{113}. The half lives were not sufficiently different, however, for the Ag\textsuperscript{112} to become neglectable before the Ag\textsuperscript{113} became too weak to measure conveniently. The two crystal method for reducing backscattering, shown in Figure IV and discussed on pages 9 to 17, was used for this determination. The source was placed directly on the coincidence crystal so no source mounting was used. The spectrum was run over twice and the points averaged to eliminate the effect of decay. The Kurie plot of the results is shown in Figure XI. An end point of 1.86 mev was obtained for the Ag\textsuperscript{113}.

The value of \( ft \) for Ag\textsuperscript{113} was calculated to be 6.1 x \( 10^6 \) seconds and (\( W_0^2 - 1 \))\( ft \) was 1.3 x \( 10^6 \) seconds, so that the spectrum may be expected to exhibit an allowed shape.
Fig. XI--Kurie plot of \( \text{Ag}^{112} \) and \( \text{Ag}^{113} \) beta-spectra.
LITERATURE CITED


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