

analyzers, an experiment of this character would not be easy.

We conclude by noting that we have considered those nuclear beta-gamma processes in which the beta transition precedes the gamma transition. The inverted transition is also possible but such transition matrix elements are not marked by the peaking at high energies in the gamma-ray spectrum in the case of near degeneracy and hence would be much more

difficult to detect experimentally. Of course many intermediate states could contribute in principle to a particular transition, but the effect of states distant in energy is relatively small.<sup>7</sup>

<sup>7</sup> On the basis of the results here presented, Dr. M. Schmorak of Brookhaven National Laboratory has undertaken a careful measurement of the gamma-ray spectrum from Ni<sup>59</sup>. A detailed analysis of his data must await completion of the calculation of the interference terms between the nuclear and electronic beta-gamma processes.

## Decay of At<sup>212</sup>; the Bi<sup>209</sup>( $\alpha, n$ )At<sup>212</sup> Reaction\*

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At<sup>212</sup> was produced by the Bi<sup>209</sup>( $\alpha, n$ )At<sup>212</sup> reaction by bombarding Bi<sup>209</sup> with a beam of 19-MeV helium ions from the Purdue cyclotron. The At<sup>212</sup> was observed to decay by alpha emission with a half-life of  $0.20 \pm 0.04$  sec. The kinetic energy of the alpha particles emitted in the decay was measured using Au-Si surface-barrier charged particle spectrometers. The alpha-decay energy was found to be  $7.87 \pm 0.07$  MeV. The relative excitation function of the Bi<sup>209</sup>( $\alpha, n$ )At<sup>212</sup> reaction was also obtained in the center-of-mass energy range from 17.87 to 18.86 MeV. The hindrance factor for the At<sup>212</sup> decay is calculated using the experimentally measured At<sup>212</sup> alpha-decay energy.

### INTRODUCTION

MANY reactions involving bismuth as the target nucleus have been studied. In particular, the ( $\alpha, 2n$ ) and ( $\alpha, 3n$ ) reactions on bismuth were studied by Kelly and Segrè.<sup>1</sup> The thresholds for the ( $\alpha, 2n$ ) and ( $\alpha, 3n$ ) reactions are at about 20 and 30 MeV, respectively. The At<sup>211</sup> and At<sup>210</sup> nuclei formed from the above reactions have half-lives of 7.2 and 8.3 h, respectively.<sup>1,2</sup> The ( $\alpha, n$ ) reaction on Bi<sup>209</sup> is more difficult to observe due to its short half-life (0.2 sec) and low cross section.

The decay of At<sup>212</sup> is interesting in that it crosses a closed neutron shell ( $N=126$ ). This hinders the decay and gives rise to a half-life which is considerably longer than would be predicted on the basis of the observed alpha-decay energy. The alpha-decay energy can be estimated from the following data: electron capture decay energy of At<sup>212</sup>  $\sim 1.8$  MeV, alpha decay energy of Po<sup>212</sup> = 8.949 MeV, and electron capture decay energy of Bi<sup>212</sup>  $\sim 2.75 \pm 0.15$  MeV.<sup>2</sup> These values give an At<sup>212</sup> alpha-decay energy of  $\sim 8.0$  MeV. Using this value in the energy-lifetime relation, a hindrance factor ( $\lambda_{ee}/\lambda$ ; where  $\lambda_{ee}$  is the partial-decay constant expected for ground-state transitions in even-even nuclei and  $\lambda$  is the

measured partial-decay constant) of  $\sim 2200$  is obtained. This is an unusually large hindrance factor and it was suggested<sup>2</sup> that either the estimated decay energy of At<sup>212</sup> was too large or the  $\frac{1}{4}$  sec half-life which had been previously measured was not the ground-state alpha decay of At<sup>212</sup>.

Two half-life measurements of At<sup>212</sup> had been made previously. In the unpublished work of Weissbluth, Putnam, and Segrè<sup>3</sup> the half-life was measured as 0.25 sec. Winn<sup>4</sup> obtained a half-life of  $0.22 \pm 0.03$  sec for the At<sup>212</sup> decay using a ZnS screen mounted on a 4-ft light pipe. No measurement of the alpha energy was made.

During the course of the present measurements, an alpha-decay energy for At<sup>212</sup> of 7.61 MeV was reported by Griffioen and Macfarlane.<sup>5</sup>

In the present experiments, Au-Si surface-barrier charged particle counters were used as detectors and energy spectrometers for the alpha particles emitted in the decay of At<sup>212</sup>. Solid-state counters were selected because they offer excellent energy resolution for alpha particles, and because with appropriate selection of the silicon resistivity and the reverse-bias applied, their response to  $\beta$ -,  $\gamma$ -, and x-ray backgrounds can be made so low that heavy particles can be detected with almost zero background even in very high background fluxes. This low sensitivity to  $\beta$ -,  $\gamma$ -, and x rays was necessary

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<sup>1</sup> E. L. Kelly and E. Segrè, Phys. Rev. **75**, 999 (1949).

<sup>2</sup> Nuclear Data Sheets, National Academy of Sciences, National Research Council (U. S. Government Printing Office, Washington, D. C.).

<sup>3</sup> M. Weissbluth, T. M. Putnam, and E. Segrè (unpublished). Reported by D. Strominger, J. M. Hollander, and G. T. Seaborg, Revs. Modern Phys. **30**, 585 (1958).

<sup>4</sup> M. M. Winn, Proc. Phys. Soc. (London) **A67**, 949 (1954).

<sup>5</sup> R. G. Griffioen and R. D. Macfarlane, University of California Radiation Laboratory Report UCRL-10023, 1962 (unpublished).

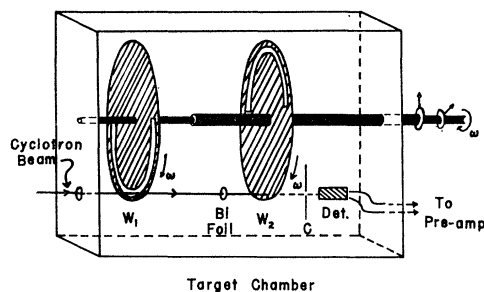


FIG. 1. Schematic drawing of target chamber used to study  $\text{Bi}^{209}(\alpha, n)\text{At}^{212}$  reaction.

since the detector was required to record the particles emitted in the  $\text{At}^{212}$  decay in the background created by the bombarding cyclotron beam.

#### EXPERIMENTAL PROCEDURES

The Au-Si surface-barrier charged particle detectors used in these experiments were constructed essentially by the procedures outlined by Blankenship and Borkowski,<sup>6</sup> and by McKenzie and Bromley.<sup>7</sup> A detailed description of the construction of these detectors is given elsewhere.<sup>8</sup> 1200  $\Omega\text{-cm}$   $n$ -type silicon<sup>9</sup> was used in the construction of the detectors. Resolutions of approximately 1½% were obtained for 8.8-MeV alpha particles using these detectors.

A solid-state detector was mounted in the target chamber as shown in Fig. 1. The target chamber was connected to the cyclotron beam tube so that the cyclotron beam passed directly into the target chamber. No separating window was required as the target chamber was maintained at cyclotron vacuum. The beam was collimated before reaching the bismuth target by a 0.250-in.-diam hole in a 0.005-in. tantalum foil. The two wheels,  $W_1$  and  $W_2$ , were rotated together at a speed of 25 rpm by a synchronous motor. A 180° circular slit in the first wheel,  $W_1$ , permitted the cyclotron helium ion beam to activate the bismuth target for ½ of each revolution (1.20 sec). The second wheel,  $W_2$ , stopped the incident beam during the time that the target was being activated. 0.03 sec after the incident helium ion beam was cut off by  $W_1$ , a slit in the second wheel,  $W_2$ , permitted the alpha particles emitted in the decay of  $\text{At}^{212}$  to pass through a 0.040-in.-diam hole in an aluminum collimator and reach the detector.

Movable  $\text{Po}^{210}$  and  $\text{Pb}^{212}$  alpha sources (5.30, 8.78, and 6.04 MeV) were included in the target chamber for calibration purposes. When not in use, the calibration sources were retracted into a compartment which was closed by a trap door in order to prevent stray counts from the calibration sources from reaching the detector.

<sup>6</sup> J. L. Blankenship and C. J. Borkowski, IRE Trans. on Nuclear Sci. 7, 190 (1960).

<sup>7</sup> J. M. McKenzie and D. A. Bromley, Proc. Inst. Elec. Engrs. 106, Pt. B Suppl. No. 16, 1959.

<sup>8</sup> J. C. Ritter, M. S. thesis, Purdue University, 1962 (unpublished).

<sup>9</sup> Kindly supplied by Merck and Company.

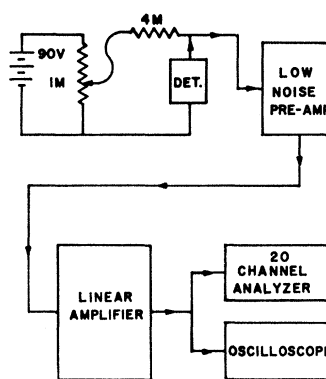


FIG. 2. Block diagram of solid-state detector electronics.

The target was prepared by vacuum evaporation of bismuth on one side of a thin (0.09 mg/cm<sup>2</sup>) gold foil. The thickness of the bismuth was measured by permitting 8.78- and 6.04-MeV alpha particles from a  $\text{Pb}^{212}$  source to pass through the target. The energy loss of the alpha particles was measured by means of the solid-state detectors. The foil thickness was then calculated using range-energy curves for lead compiled by Whaling.<sup>10</sup>

A block diagram of the electronics is given in Fig. 2. The charge sensitive, low-noise preamplifier was constructed from a design reported by Fairstein.<sup>11</sup>

The energy of the alpha particles emitted by  $\text{At}^{212}$  was measured in the following way. The target chamber (see Fig. 1) was connected to the exit port of the cyclotron so that the 1.7 mg/cm<sup>2</sup> bismuth foil was bombarded by an unanalyzed beam of helium ions from the cyclotron. The energy range of the incident beam was approximately 18½–19½ MeV. The 180° slit in the rotating wheel permitted the beam to activate the bismuth target until secular equilibrium of the  $\text{At}^{212}$  activity was nearly reached. After the helium ion beam was cut off by the first wheel, the slit in the second wheel permitted the alpha particles from the  $\text{At}^{212}$  decay to reach the detector for approximately five half-lives. The cycle was repeated continuously until sufficient counts had been accumulated. The linearity of the electronics was checked immediately prior to running by means of a precision mercury relay pulser. Before and after each run, the entire system was also calibrated using the  $\text{Po}^{210}$  and  $\text{Pb}^{212}$  alpha sources. The calibration curves taken before and after each run never differed in energy by more than 0.3%.

The observed peak due to the alpha decay of  $\text{At}^{212}$  is shown in Fig. 3. This peak was obtained by summing four separate runs. A calibration curve obtained using  $\text{Po}^{210}$  and  $\text{Pb}^{212}$  alpha sources as well as the pulser linearity check is also shown. Three types of background were taken. In the first, both wheels were stopped and the bombarding beam was allowed to

<sup>10</sup> Ward Whaling, *Handbuch der Physik*, edited by S. Flugge (Springer-Verlag, Berlin, 1957), Vol. 28.

<sup>11</sup> J. L. Blankenship and C. J. Borkowski, IRE Trans. on Nuclear Sci. 8, 17 (1961).

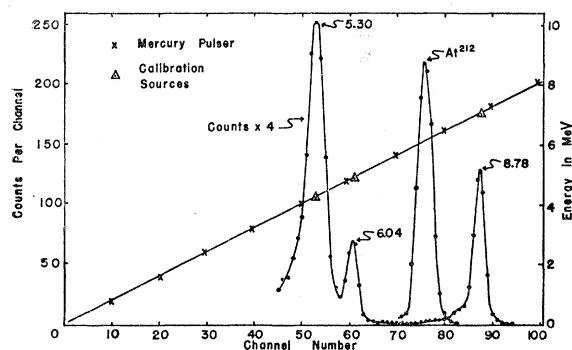


FIG. 3. Observed spectrum from the alpha decay of  $\text{At}^{212}$ . Spectra from 8.78-, 6.04-, and 5.30-MeV calibration sources and pulser linearity check of the electronics are also shown.

strike the first wheel, while the slit in the second wheel opened the path from the bismuth target to the detector. The second was taken with wheels  $W_1$  and  $W_2$  rotating and the bombarding beam on, but the Au-backed bismuth target was replaced with a similar gold backing with no bismuth deposit on this target. The third run was taken with the gold-backed bismuth target; the run was started 10 min after the cyclotron was turned off following a full day of operation. In all background runs no appreciable count rate was observed in the region of the  $\text{At}^{212}$  decay peak. No peaks other than the  $\text{At}^{212}$  decay peak were observed. Any peak corresponding to an alpha energy above 2 MeV and with a counting rate greater than about 10% that of the  $\text{At}^{212}$  decay peak would have been observed.

In the half-life measurement, the circuit shown in Fig. 4 was used. The microswitch was activated by a cam which rotated with the wheels,  $W_1$  and  $W_2$  described above. During most of each revolution the spectrum was displayed in channels 1–10 of a 20-channel analyzer. As the wheels rotated, the cam activated the micro-switch during a small portion of the cycle. During the time the microswitch was activated, the mercury relay added the lower 50- $\Omega$  resistor (which was normally shorted) to the circuit, thus switching the spectrum from the first ten channels to all 20 channels of the analyzer.

By rotating the cam to various angles with respect to the opening in the wheel  $W_2$ , any portion of the decay could be selected and the peak displayed in the upper channels.

The relative fraction of the time per cycle that the microswitch was activated was determined at each position of the cam used by repeating the above measurement with a  $\text{Po}^{210}$  source placed immediately behind the bismuth foil. During this measurement the bismuth foil was not activated by the cyclotron beam, but the wheels were rotated. This measurement was performed both before and after each  $\text{At}^{212}$  half-life measurement.

The  $\text{At}^{212}$  counts obtained in channels 10–20 per unit of time the microswitch was activated represented the count rate which occurred during the selected portion of

the decay. The relative counting rates, corrected for the microswitch activation times, were plotted as a function of elapsed time after the bombardment ceased. The curve was fitted by the method of least squares and the half-life was determined from the slope of the line of best fit.

In order to obtain the relative cross section as a function of the energy of the incident helium ions for the  $\text{Bi}^{209}(\alpha, n)\text{At}^{212}$  reaction, a 1.7 mg/cm<sup>2</sup> bismuth target on a 0.09 mg/cm<sup>2</sup> gold backing was bombarded with an energy-analyzed beam. The lab energy of the bombarding beam was varied from 18.25 to 19.25 MeV. The beam which passed through the target was collected by wheel  $W_2$  and was integrated by a current integrator. The current integrator was calibrated after the experimental measurements were completed using a constant current source. It was assumed that the current integrator would integrate properly the pulsating dc applied during the experiments, and that secondary electron emission from

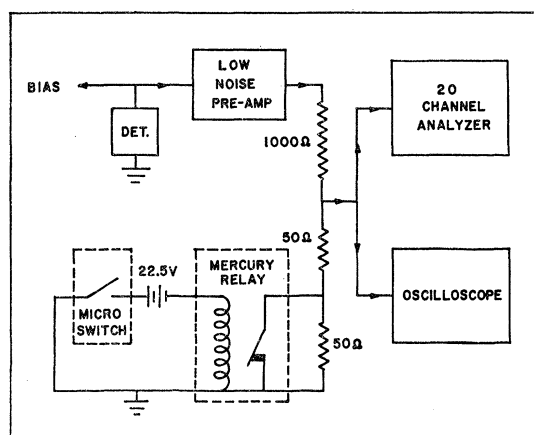


FIG. 4. Block diagram of electronics for  $\text{At}^{212}$  half-life measurement.

$W_2$  was constant over the incident beam energy range. The rms energy spread of the beam at 19.00 MeV was  $\sigma_{\text{rms}} = 32$  keV. The absolute error associated with the beam energy determination was 0.1%.

## EXPERIMENTAL RESULTS

The alpha particles emitted in the decay of  $\text{At}^{212}$  peaked at a pulse height corresponding to an energy of  $7.52 \pm 0.05$  MeV. This value must be corrected for the energy loss of the alpha particles emitted by the  $\text{At}^{212}$  as they traverse the remainder of the bismuth foil from their point of origin to the detector. Due to the energy dependence of the cross section of the  $\text{Bi}^{209}(\alpha, n)\text{At}^{212}$  reaction, more  $\text{At}^{212}$  nuclides are formed at the back side of the bismuth foil than at the detector side. An un-analyzed helium ion beam was necessary in order to obtain reasonable counting rates for the energy measurement. In order to approximate the number of  $\text{At}^{212}$  atoms formed in the bismuth foil as a function of dis-

tance from the back of the bismuth foil, it was assumed that the incident beam was monoenergetic with a lab energy of 19.00 MeV. Using range-energy curves compiled by Whaling<sup>9</sup> and the experimentally observed relative  $\text{Bi}^{209}(\alpha, n)\text{At}^{212}$  excitation function, the relative number of  $\text{At}^{212}$  atoms produced as a function of distance from the back of the gold-bismuth target was calculated.

The 1.7 mg/cm<sup>2</sup> bismuth foil was imagined to be divided, parallel to the foil faces, into a series of thin sections of equal thicknesses. The relative number of  $\text{At}^{212}$  atoms formed in each section was then calculated. The alpha emission contributed by each section of the foil was approximated in the following way. Consider a plane source of alpha particles located at the center of each of the thin sections of the foil. The response of the detector to one of these alpha sources would be a

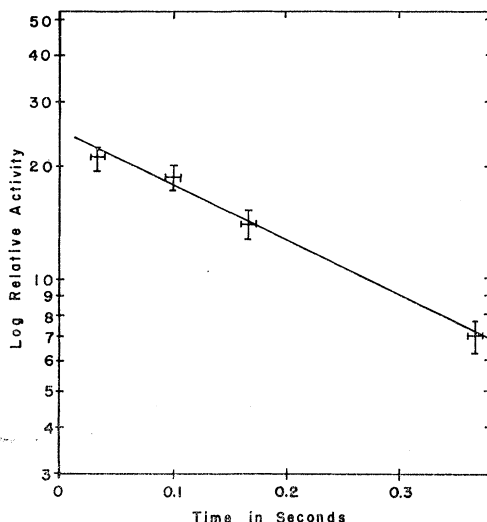


FIG. 5. Semilogarithmic plot of relative counting rate of  $\text{At}^{212}$  alpha particles as a function of time. The curve was fit by the method of least squares.

Gaussian-shaped spectrum peaked at the energy  $E_\alpha - \Delta E_\alpha$  where  $E_\alpha$  is the energy of the alpha particles emitted by the source and  $\Delta E_\alpha$  is the average energy lost by the alpha particles in traversing the thickness of foil between the source and detector. The half-width of each Gaussian was assumed to be the same as that observed for the 8.78-MeV calibration peak since the experimental half-width was very nearly constant from 8.78 to 6.04 MeV. The height of the Gaussian from each section of the foil was made proportional to the number of counts originating in that section. The calculated response of the detector to all the line sources was formed by summing the counts from each source at a series of energies. The resulting sum peak thus formed was compared to the observed  $\text{At}^{212}$  peak. The full width at half-maximum of the calculated peak was 4.7% which is in good agreement with the observed full

width at half-maximum of 4.8%. The peak of the sum curve occurred at  $7.72 \pm 0.07$  MeV which corresponds to the kinetic energy of the alpha particles emitted by  $\text{At}^{212}$  including the correction for target thickness. Including the recoil energy of the daughter nucleus, the alpha-decay energy of  $\text{At}^{212}$  is  $Q_\alpha(\text{At}^{212}) = 7.87 \pm 0.07$  MeV.

The half-life calculated from a least squares fit of the experimental data was  $T_{1/2} = 0.20 \pm 0.04$  sec (see Fig. 5). This value agrees, within the experimental errors, with the value of  $0.22 \pm 0.03$  sec obtained by Winn.<sup>4</sup>

It should be noted that the present measurements uniquely correlate the 0.22-sec half-life with a 7.87-MeV alpha transition, since only one alpha group was observed in the experiment.

The relative cross section as a function of energy for the  $\text{Bi}^{209}(\alpha, n)\text{At}^{212}$  reaction is shown in Fig. 6. Corrections to this curve for target thickness were negligible. A theoretical curve plotted from the data of Blatt and Weisskopf<sup>12</sup> is shown on the same graph for comparison. The theoretical curve was normalized to the experimental curve at a center-of-mass energy of 18.6 MeV. The theoretical curve was obtained from the continuum theory of nuclear reactions for  $r_0 = 1.5 \times 10^{-13}$  cm where  $r_0$  is the nuclear radius parameter. This theoretical curve represents the cross section for formation of the compound nucleus, C, where  $\text{Bi}^{209} + \text{He}^4 = \text{C}$ , by bombardment of  $\text{Bi}^{209}$  with helium ions. If a reaction with neutron emission is energetically possible, then the cross section for the  $(\alpha, n)$  reaction is approximately equal to the

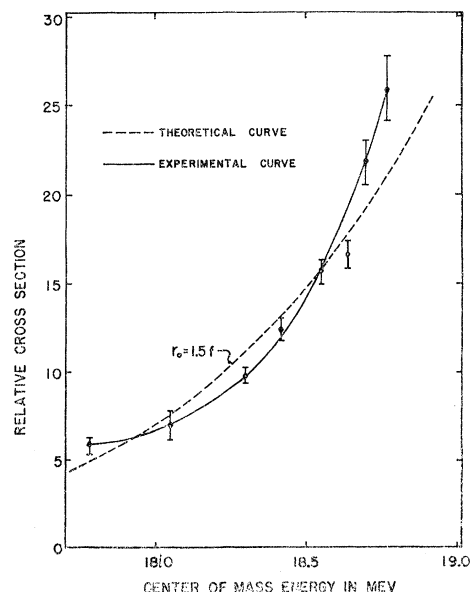


FIG. 6.  $\text{Bi}^{209}(\alpha, n)\text{At}^{212}$  excitation function. A theoretical curve for  $r = 1.5 \text{ F}$  is also shown. The theoretical curve is plotted from the data of Blatt and Weisskopf<sup>12</sup> and is normalized to the experimental curve at 18.6 MeV.

<sup>12</sup> J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics* (John Wiley & Sons, Inc., New York, 1952), p. 353.

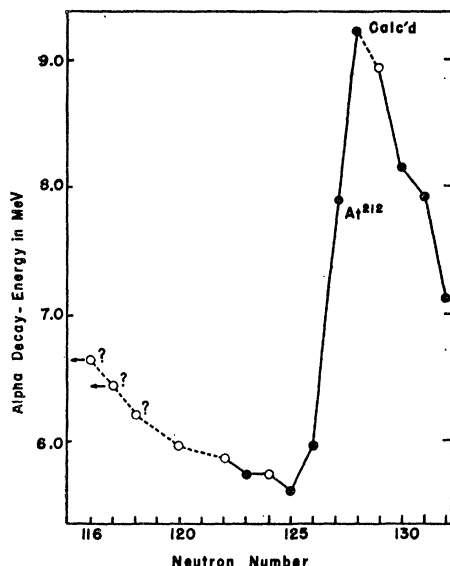


FIG. 7. Alpha-decay energy of astatine isotopes as a function of neutron number. Data (except for  $\text{At}^{212}$  point) taken from Hanna (see reference 15.) The open circles indicate uncertainty in the mass number assignment.

cross section for formation of the compound nucleus until the threshold (about 20 MeV) for the  $(\alpha, 2n)$  reaction is reached. The value obtained for the absolute cross section at 18.6 MeV was  $15.5 \pm 3$  mb; this is a lower limit since no provision was made to suppress secondary electron emission from the beam collecting wheel.

### DISCUSSION

The presently determined alpha-decay energy of  $\text{At}^{212}$ ,  $7.87 \pm 0.07$  MeV, may be compared with the result of Griffioen and Macfarlane,<sup>5</sup> 7.61 MeV; and with the estimated value (based on other experimental results and nuclear systematics) of  $\sim 8.0$  MeV<sup>2</sup>.

Griffioen and Macfarlane do not give an error with their result. However, it is probably less than the presently quoted error of 0.07 MeV because they do not have to correct their result for source thickness. The

difference between the two results is 2 to 3 times as large as the sum of the errors of the measurements. No reason for this discrepancy is apparent.

The present 7.87-MeV value is in agreement, within the uncertainties involved, with the estimated value of  $\sim 8.0$  MeV.

$\lambda_{ee}$  was calculated from the semiempirical relation:

$$\log_{10} T_{1/2} = A(Q_{\alpha})^{-1/2} + B,$$

where  $A = 133.40$  and  $B = -51.1913$  for astatine.<sup>13</sup> The constants were obtained by interpolation between neighboring even- $Z$  values. Using the presently determined value for the alpha-decay energy of  $\text{At}^{212}$ , 7.87 MeV, a hindrance factor of  $\sim 900$  was obtained. Here it has been assumed that the measured alpha half-life of  $\text{At}^{212}$  is the partial half-life of the decay. It is also assumed in the calculation of  $Q_{\alpha}(\text{At}^{212})$  that the decay occurs from the ground state of  $\text{At}^{212}$  to the ground state of  $\text{Bi}^{208}$ . This is consistent with the other experimental data used in the estimation of  $Q_{\alpha}(\text{At}^{212})$ . Perlman *et al.*<sup>14</sup> have suggested that large hindrance factors are due, at least partly, to an unusually small preformation probability for the alpha particle from unpaired nucleons.

Figure 7 shows the variation in alpha-decay energy of astatine isotopes as a function of neutron number. The data for the curve except for the  $\text{At}^{212}$  point were taken from Hanna.<sup>15</sup> The large decrease in alpha-decay energy at the  $N=126$  closed shell is due to the fact that a neutron must be removed from within the closed  $N=126$  shell in order to form the alpha particle for the decay.

### ACKNOWLEDGMENTS

The authors wish to thank Dr. E. Bleuler for his suggestion of the electronics for the half-life measurement, and C. L. Wiley, J. W. Hemskey, and S. S. Wong for help in making the foil thickness measurements. Acknowledgment is also made to E. L. Robinson for helpful discussions.

<sup>13</sup> G. C. Hanna, *Experimental Nuclear Physics*, edited by E. Segre (John Wiley & Sons, Inc., New York, 1959), Vol. 3, p. 116.

<sup>14</sup> I. Perlman, A. Ghiorso, and G. T. Seaborg, *Phys. Rev.* **77**, 26 (1950).

<sup>15</sup> G. C. Hanna, reference 13, p. 68.