The foregoing discussion is summarized in an energy-level diagram of N^{14} (Fig. 5) showing the established levels below 10 Mev. Besides the levels discussed above, there are two additional ones at 7.0 Mev and 7.7 Mev reported in reference 19. Since guesses at the probable types of radiation were made in making some of the angular momentum assignments shown in Fig. 5, one cannot uniquely confirm theoretical estimates of transition probabilities for the different types of radiation. However, further work to determine the complete angular distributions of the many gamma rays found at these several proton resonance levels promises to be a fruitful method for uniquely determining the angular momentum of the levels and the types of radiation involved in N^{14}.

Since this work has extended over a period of two years, it is impossible to list individually all those who aided directly or indirectly in the taking of data or in the interpretation of the results. It is a pleasure, however, to acknowledge the contributions and continued interest of Professor C. C. Lauritsen and the staff of the Kellogg Radiation Laboratory.

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Formation of the Long-Lived Bi^{210} Isomer*

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The thermal activation cross section of Bi^{209} is shown to be definitely less than the absorption cross section, 19 ± 2 mb as compared with 33 ± 2 mb. In order to verify the high absorption value, previously obtained by pile reactivity methods, measurements were made with the long wavelength neutrons available with the Brookhaven slow chopper. The absorption cross section is obtained from the chopper measurements by subtraction of thermal inelastic scattering. The latter scattering is determined from the variation of measured scattering with sample temperature. The difference between the absorption and scattering, or 14 ± 3 mb, is ascribed to the formation of the long-lived α-emitting isomeric state of Bi^{210}, leading to a half-life of $(2.6\pm0.8)\times10^6$ years. This long life is in agreement with the expected properties, relative to α and β decay, of a high-spin state of Bi^{210}.

I. INTRODUCTION

THE absorption cross section of bismuth for thermal neutrons is extremely small, constituting only a fraction of a percent of the scattering cross section. Early measurements[1] of the activation cross section, i.e., the cross section for activation of the five-day Bi^{210} (RaE), resulted in a value of about 15 mb. On the other hand, the absorption cross section, which is measured by some phenomenon sensitive to the disappearance of neutrons, gave somewhat higher values, of the order of 30 mb. However interfering effects of the large scattering cross section in the absorption measurements, as well as the possible contribution of impurities, made it seem likely that the true absorption could easily be the same as the activation cross section, i.e., about 15 mb.

It soon became clear that the difference between absorption and activation was not a result of impurities because the preparation of samples of increasing purity, as evidenced by chemical analysis, did not give rise to any decrease in absorption cross sections. The measured values lay in the 30–35-mb range regardless of the sample origin and degree of purification. In 1948, measurements of bismuth absorption were made by Langsdorf[2] with the "pile oscillator," and his value for pure bismuth was again about 30 mb. However, because of the interfering effects of scattering in the pile, it was felt by Langsdorf that some apparent absorption could result from neutron scattering and that the correct absorption of bismuth, after correction for this scattering effect, could be as low as the activation value.

A series of measurements of the absorption and activation cross sections was carried out by Palevsky, Hughes, and Eggler[3] in 1950 to investigate the possibility of a real difference between these cross sections, the experiments being performed in the graphite pile to study the effect of scattering on the absorption cross section. These measurements showed that there was indeed a distinct difference between the cross section for activation of the 5-day RaE and the absorption, indicating the formation of a hitherto undetected activity in bismuth in addition to the well-known five-day period. At about the same time a long-lived α-emitting activity was found in pile-irradiated bismuth by Newman, Howland, and Perlman,[4] and it seemed quite likely that the difference between absorption and activation could be attributed to the formation of the long-lived state. To investigate this possibility, as well

* Work carried out under contract with the U. S. Atomic Energy Commission.
[1] Seren, Friedlander, and Turkel, Phys. Rev. **72**, 888 (1947).
[2] A. S. Langsdorf, Phys. Rev. **74**, 1217 (1948).
[3] Palevsky, Hughes, and Eggler, Phys. Rev. **83**, 234 (1951).
[4] Newman, Howland, and Perlman, Phys. Rev. **77**, 720 (1950).

as to determine the spin-dependent scattering needed in connection with experiments on the neutron-electron interaction,[5] additional work on the thermal cross sections of bismuth has been carried out. This work has included "cold" neutron measurements made with the Brookhaven slow chopper.[6] In the present paper the earlier results will be reviewed briefly, the recent ones given in more detail, and the lifetime of the α-emitting state estimated.

II. THERMAL NEUTRON RESULTS

The activation cross section, while inherently inaccurate to about ten percent, is simple to perform and contains very little chance of gross misinterpretation. As the cross section for the formation of an activity of a particular decay period is measured, there is small possibility of contribution from an impurity. Errors of several percent arise, however, because of the difficulty of flux standardization and disintegration rate measurement of the activated sample.[7] The results that have been obtained for the activation cross section of the five-day period show some variation, but their essential agreement justifies the conclusion that there is no large error in this cross section. The value obtained by Seren[1] was 15 ± 3 mb, that of Palevsky, Hughes, and Eggler[3] was 17 ± 3 mb, and recent measurements at Harwell[8] gave the slightly higher result of 20.5 mb with an error of only 1.1 mb. The "best value" given in the Atomic Energy Commission cross-section compilation[9] is 19 ± 2 mb, which we shall adopt in this work in spite of the larger stated error relative to the Harwell result.

The two common methods of measuring the absorption cross section, usually referred to as the "danger coefficient" and the "pile oscillator" methods, give the neutron absorption in terms of the effect on the reactivity of a pile. In the first method a sample is placed inside the pile, and the reactivity is then determined by adjustment of a control rod; in contrast to this static approach, the sample is oscillated in the second method, and the oscillating component of the pile power is a measure of the neutron absorption. The effect of neutron scattering in causing an apparent absorption in the oscillator method arises because the sample scatters neutrons out of the pile to an extent depending on its position and this removal of neutrons affects the pile reactivity. In the danger-coefficient method, the central position of the absorbing sample means that its scattering has practically no effect. A second scattering effect, thought by Langsdorf[2] to cause a larger apparent absorption in bismuth, consists of a lowering of the neutron energy by inelastic scattering in bismuth so that the neutrons no longer cause fission in U^{238}, resulting in a loss in reactivity that would appear as absorption.

In the measurements made by Palevsky, Hughes, and Eggler[3] in the graphite pile, the possibility of the effect suggested by Langsdorf was checked by variation of the geometrical relationship of the sample to the uranium fuel lumps. The absorption cross section was measured by the standard danger-coefficient method utilizing two arrangements, in one of which all of the uranium fuel lumps were removed from the vicinity of the sample, whereas in the other the fuel lumps were in close proximity to the sample. As these measurements gave no differences in the measured absorption cross section for the two arrangements, it was clear that there was no appreciable effect of inelastic scattering on reactivity. In the danger-coefficient work a number of samples of the highest purity that could be attained were measured, and an extremely small variation in absorption cross section was observed from sample to sample; in fact, most of the samples tested, which had received various degrees of chemical purification, were in the range 30–35 mb. The final value obtained was 33 ± 2 mb, with no correction for absorption by impurities because chemical analysis revealed none that would contribute significant absorption.

The results of these measurements thus seemed to indicate very strongly that the absorption was actually of the order of twice the activation cross section and furthermore that the difference could be attributed to the formation of the long-lived α-emitted state. However, because of the interest in the use of bismuth for the neutron mirror experiment as well as for the definite settlement of the thermal cross section, experiments with the Brookhaven slow chopper were carried out as well.

III. COLD-NEUTRON MEASUREMENTS

In the determination of the absorption cross section of bismuth, a total cross-section measurement made by transmission is of no value when this measurement is made at thermal energies because the scattering is so much larger than the absorption cross section. However for a total cross-section measurement made at long wavelengths (beyond the crystal "cutoff"), the coherent scattering does not appear in a transmission measurement, and the absorption cross section can usually be identified with certainty.[10]

The coherent scattering "disappears" (in actuality it is only forward and hence not observed) at a wavelength equal to twice the longest lattice spacing of the crystal. For longer wavelengths the observed cross section consists only of capture plus elastic incoherent scattering (spin-dependent and isotopic) and inelastic

[5] Hughes, Harvey, Goldberg, and Stafne, Phys. Rev. **90**, 497 (1953).
[6] Seidl, Palevsky, Randall, and Thorne, Phys. Rev. **82**, 345 (1951).
[7] D. J. Hughes, *Pile Neutron Research* (Addison-Wesley Press, Cambridge, 1953), pp. 179–188.
[8] D. J. Littler and E. E. Lockett (private communication).
[9] *Neutron Cross Sections*, Atomic Energy Commission Report AECU-2040-Supplement 2 (Office of Technical Services, Department of Commerce, Washington, D. C., 1952).
[10] See reference 7, pp. 249–254.

lattice-vibration scattering. The elastic incoherent scattering shows no variation with wavelength or sample temperature and the inelastic scattering varies rapidly with temperature and is proportional to wavelength at large wavelength, whereas the capture cross section is usually proportional to wavelength and is always independent of sample temperature. Investigation of the total cross section at wavelengths beyond cutoff as a function of wavelength and sample temperature thus makes it possible to identify the various components of the incoherent scattering.[10]

For certain materials some components of the incoherent scattering are zero, and the analysis is correspondingly simplified; for example, an element of zero nuclear spin (an even-even isotope, such as O^{16}, U^{238}) produces no spin-dependent scattering, and no isotope incoherence results for a monoisotopic element. While it is true that all materials show some inelastic scattering, calculations[11] based on the Debye frequency spectrum of lattice vibrations usually predict the inelastic scattering with reasonable accuracy.

In the case of monoisotopic bismuth no isotopic incoherence will be present, but, as the spin is 9/2, spin-dependent incoherence is possible. The thermal inelastic scattering could be reduced to a negligible value by cooling the sample to a few degrees absolute, and the observed total cross section would then consist only of 1/v capture and spin-dependent incoherence. However, sufficient accuracy can be obtained by cooling the bismuth to only liquid-air temperature (i.e., about 100°K). The absorption and the spin-dependent scattering can be determined by measurements of the total cross section for different sample temperatures and extrapolation to remove the inelastic scattering still present at 100°K. The inelastic scattering itself is of course also measured in the process, but as it is not of direct interest in the absorption discussion, it will not be considered in detail.

The total cross sections for cold neutrons were measured with the Brookhaven slow chopper,[6] which has been used for several years in the long-wavelength region for measurements of inelastic,[12,13] spin-dependent,[14] and absorption cross sections.[15] We shall not describe the techniques used with the chopper for control of fast-neutron background by filters, for background determination and for wavelength standardization; these matters are covered in reference 7, pp. 243–249, and in reference 15. The results for room temperature bismuth are shown in Fig. 1, in which the total cross section is compared with the theoretical inelastic scattering.[11]

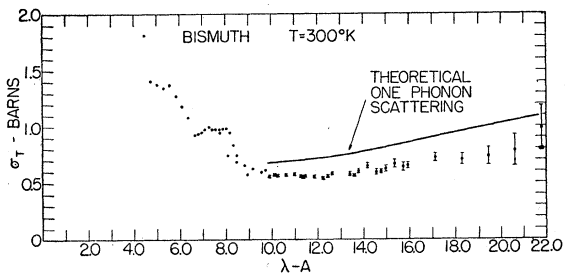

FIG. 1. The measured total cross section of room temperature bismuth compared with the theoretical inelastic scattering in which gain or loss of single phonons alone is considered.

The inelastic scattering in bismuth is unusual in that it exhibits a flat minimum in the region of the cutoff wavelength (about 8A) and then gradually approaches the 1/v or "λ law" region. The room-temperature results exhibit the expected wavelength behavior of the inelastic scattering but cannot be used to obtain a value for the 1/v capture cross section because the observed *total* cross section is less than the theoretical inelastic scattering alone. Because of the failure of the theory to predict the absolute magnitude of the inelastic scattering,[16] it is necessary to investigate the variation of cross section with temperature in order to identify the absorption.

In order to investigate the temperature variation of the inelastic scattering, measurements were made at high sample temperature where the inelastic scattering is the major component of the cross section. In these measurements the bismuth sample was contained in a furnace at a temperature of 500°K, controlled to a few degrees. The results of cross-section measurements at 500°K are shown in Fig. 2, again compared with the theoretical inelastic scattering. At this temperature the inelastic scattering is large, and, as it is the major component of the scattering, it can easily be compared with the theoretical value. Although the experimental points are only about 50 percent of the calculated values (using the danger-coefficient results to correct the former for absorption), the increase at long wavelength

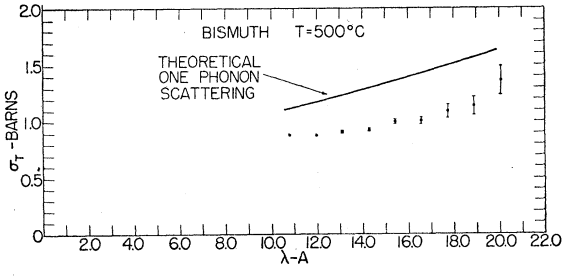

FIG. 2. The measured total cross section of bismuth at 500°K compared with theory.

[11] Carried out for bismuth by D. Kleinman, Phys. Rev. **81**, 326 (1951), and by G. Placzek and L. Van Hove (unpublished).

[12] Palevsky, Hughes, and Smith, Phys. Rev. **87**, 221 (1952).

[13] Johnson, Palevsky, and Hughes, Phys. Rev. **82**, 345 (1951).

[14] H. Palevsky and R. R. Smith, Phys. Rev. **86**, 604 (1952).

[15] Carter, Palevsky, Meyers, and Hughes, Phys. Rev. **92**, 714 (1953).

[16] This "failure" results from an assumption in the theory that waves scattered from different nuclei do not interfere (the "incoherent approximation").

(the λ law) is about as predicted by theory. Fortunately, in the actual extrapolation used to obtain the inelastic scattering at low temperature, only the form of the temperature variation, not the absolute magnitude of the theoretical result, need be used.

For the low-temperature measurements, the sample was placed in a refrigerator in which it could be moved into and out of the beam by remote control and kept at the same temperature (100°K) in both positions. Sturdy construction was necessitated by the large samples; a sample length of 15 in. gave a transmission of 0.7 at the minimum cross section. Careful checks were made to demonstrate that the sample would not affect the open beam intensity when it was out of the beam. The experimental points as well as the theoretical contribution of the inelastic scattering are given in Fig. 3. The flat minimum in the cross section indicates that the inelastic scattering, although small, exhibits the correct wavelength behavior.

In order to obtain the 1/v absorption cross section without use of the calculated absolute value of the inelastic scattering, the measured cross sections can be extrapolated to a temperature where the inelastic contribution is zero. Above 100°K the inelastic scattering is expected[11] to vary almost linearly with temperature, following a line that, *extrapolated*, intersects the temperature axis at 36°K, although the actual cross section below 100°K remains finite. The removal of the inelastic scattering by extrapolation in this manner is exhibited in Fig. 4, in which the observed total cross sections are plotted against temperature for several wavelengths. These long wavelengths were chosen to accentuate the absorption relative to inelastic and spin-dependent scattering. A small theoretical correction (a 7-percent reduction) has been applied to the 100°K points because of a departure from the linear behavior caused by scattering in which the neutrons *gain* energy. The linear extrapolations to 36°K give cross sections consisting primarily of absorption, together with a small spin-dependent contribution. The magnitude of the inelastic scattering present at 100°K, which is removed by extrapolation, is given by comparison of the linear cross-section curve at 36°K and 100°K and is seen to be relatively smaller for long wavelengths.

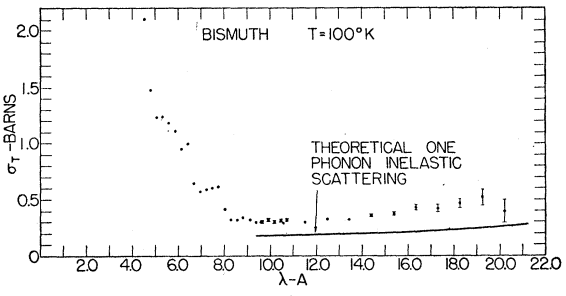

FIG. 3. The total cross section of low temperature (100°K) bismuth and the theoretical inelastic scattering.

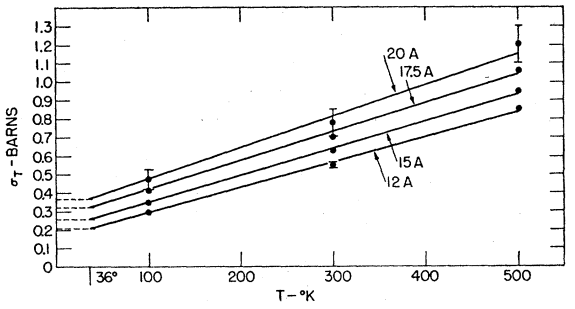

FIG. 4. The observed total cross sections for several wavelengths as functions of temperature. The linear extrapolation to 36°K is used to eliminate the inelastic scattering.

The cross sections determined by extrapolation in Fig. 4 must be corrected for the relatively small (at long wavelength) incoherent scattering before the 1/v absorption can be obtained. The separation of the extrapolated cross sections into these two components is illustrated in Fig. 5, in which are given the results of Fig. 4 as a function of wavelength. The intercept on the cross-section axis in Fig. 5 gives a spin-dependent cross section of 0±20 mb and the slope a 1/v absorption of 33±5 mb at 1.80A (2200 meters/sec).

The spin-dependent scattering obtained in Fig. 5 is extremely small, constituting only 0.2 percent of the scattering cross section at the upper limit of error. As the bismuth nucleus consists of a closed cell plus one proton, it might be expected that the interaction of the neutron, if primarily with the odd proton, would be largely spin-dependent. The extremely small spin-dependence found seems to imply that the interaction takes place with the entire nucleus rather than with the odd proton.

The absorption cross section resulting from the cold-neutron measurements is free from any corrections of the type that affect the pile oscillator and danger-coefficient techniques, although in this case of less accuracy. This constitutes convincing evidence that the absorption cross section is actually 33 mb, definitely higher than the 19-mb absorption cross section. The

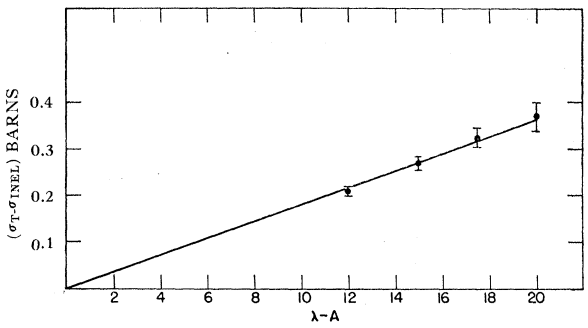

FIG. 5. The extrapolated cross sections of Fig. 4 as a function of wavelength. The slope of the curve gives the 1/v absorption (33±5 mb) and the intercept gives the incoherent elastic (spin-dependent) scattering (0±20 mb).

bismuth used for the cold neutron measurements was carefully checked for impurities by chemical analysis and was also measured relative to other high purity samples by the danger-coefficient technique. No impurities that would contribute significant absorption were detected, and the danger-coefficient results were the same within experimental uncertainty (±2 mb) as for the bismuth of highest purity used in the earlier danger-coefficient work.

IV. CONCLUSIONS

The fact that the cold-neutron measurements give the same absorption for bismuth as the danger-coefficient results shows that the interfering effect of scattering has been successfully eliminated and that the results represent actual neutron absorption. That the absorption is in bismuth itself and not in impurities is indicated by the finding that the cross section as measured by pile reactivity does not decrease with continued chemical purification once the cross section is in the 30–35-mb range. Actual chemical analysis of the various samples used in the danger coefficient as well as in the cold neutron measurements verifies that there is no appreciable absorption contributed by impurities. The impurities in the bismuth samples used by Littler and Lockett[8] constituted about 7-mb absorption, and their absorption result, which is 30.8±2.2 mb after correction for the impurities, is in good agreement with ours (33±2 mb), containing no appreciable impurity contribution. It thus seems certain that the difference between the absorption cross section (33±2 mb) and the activation cross section (19±2 mb), or 14±3 mb, must result in the formation of an activity in Bi^{210} different from the five-day RaE.

The isomeric bismuth activity seems certain to be the long-lived alpha emitter found by Newman, Howland, and Perlman[4] in highly irradiated bismuth. The isomeric activity was shown by Newman *et al.* to have a half-life greater than 25 years, fixed by the failure to decay, and to have a half-life for β or γ emission of greater than 5×10^4 years, determined by the lack of growth of the corresponding daughter activities. The present results allow a calculation of the half-life of the α-activity, a half-life much longer than the lower limit of 25 years. Newman *et al.* observed that their sample had a specific activity of 1.9±0.1 α disintegrations per minute per mg. This specific activity, I, together with the formation cross section of 14 mb, then gives the half-life in terms of the flus nv and irradiation time t,

$$T_{\frac{1}{2}}=0.693nvN\sigma t/I,$$

where N is the number of Bi nuclei per mg. The quantities nv and irradiation time t were not given by Newman *et al.*, but the product nvt, which is known[17] from the Po^{210} α activity of the sample, gives for the half-life the value,

$$T_{\frac{1}{2}}=(2.6\pm0.8)\times10^6 \text{ years}.$$

The uncertainty in this result arises principally from the quantity nvt and the cross section for formation of the isomeric state. The conclusion is certain, however, that the half-life is extremely long, and much greater than the directly observed lower limit.

There is still some question concerning the location of the α-emitting energy level, although the most recent energy values[18,19] indicate that it is 28±26 kev *below* the 4.85-day state. Should this order of energy levels be correct, the difficulty[20] is removed that was presented by the reverse order, i.e., the long half-life of the α-emitting state toward γ emission. Actually the long half-life obtained in the present work constitutes a solution to the additional problem, discussed by Feather,[20] of the failure of Bi^{210} to fit the Geiger-Nuttall diagram for $Z=83$. Whereas the 100-year half-life used by Feather disagreed seriously with the diagram, the present value gives good agreement. Actually the failure of the α-emitting state to decay by β emission to Po^{210} and the extremely long α half-life both require high spin for this state of Bi^{210}, of the order of 6.

The possibility of levels in Bi^{210}, of widely different spins and small energy separation, has been discussed by Pryce[21] in terms of the single-particle model. He has estimated the energies for different configurations of the neutron and proton moving in the field of the closed shell, taking into account their mutual interaction, and in this way has made plausible the existence of levels of the type needed to explain the properties of the bismuth isomers.

We wish to express our thanks to C. Eggler, of Argonne Laboratory, for his assistance in checking the purity of samples and to D. J. Littler and E. E. Lockett for a prepublication copy of their paper.

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[18] H. B. Levy and I. Perlman, Phys. Rev. **85**, 758 (1952).
[19] D. E. Alburger and G. Friedlander, Phys. Rev. **82**, 977 (1951).
[20] N. Feather, Phil. Mag. **42**, 568 (1951).
[21] M. H. L. Pryce, Proc. Phys. Soc. (London) **65**, 773 (1952).