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# Interference Phenomena in the Scattering of Slow Neutrons*

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Transmission measurements of the neutrons absorbed by cadmium (C neutrons) are described which show that the total cross sections for various elements are dependent to a marked degree upon the physical and chemical state of these elements. This is shown by (1), the transparency of single crystals as compared with the same material in a polycrystalline form, and (2), the nonadditivity of the cross sections for chemical compounds and alloys. The de Broglie wave-length of the C neutrons is such that the above effects can be interpreted in terms of interference. The nature of the results is well illustrated by the $SiO_2$ measurements. The cross section for the molecule computed from measurements on the component elements is 10.5. The cross section for the molecule in sand is $8.8 \pm 1$; in fused quartz $8.1 \pm 1$; and in single crystals of quartz $4.3 \pm 0.6$, all in units of $10^{-24}$ cm$^2$.

WE[1-3] have studied experimentally the modifications in the slow neutron interaction which result from changes in the physical and chemical state of the materials with which the neutrons are interacting. The wave-lengths of the neutrons used are of the right order of magnitude for these modifications to arise from interference effects in the coherently scattered portion of the neutron beam.[4]

Several theoretical[4-7] discussions of the interference of slow neutrons have been given. It has been pointed out[4, 6] that in the case of the scattering of neutrons there may be important modifications resulting from relative phase shift differences when more than one type of nucleus is present. The amplitude and phase shift will in general be different for each nucleus and for each isotope and possibly depend on the spin orientation of the nucleus. It should be pointed out that the phase shifts which occur for slow neutrons are close to $N\pi$ where $N$ is an integer. This is necessary as the cross section for the scattering of slow neutrons is small compared with $\lambda^2$. The existence of Bragg reflection at the Bragg angle from a crystal composed of two nuclei, with comparable scattering cross sections, would show that the phase shifts in the scattered waves from the two nuclei are of the same class, i.e., $N$ is even or odd for both of them. By such experiments scattering nuclei may be put into two classes one having even values and one having odd values of $N$.

Preiswerk and Von Halban[8] have attempted to observe the diffraction of slow neutrons. Their experiment consisted of a comparison of the angular distribution of 90°K and 300°K neutrons scattered from a cylinder of iron. They considered the observed change in angular distribution to be in qualitative agreement with that expected from calculations of Elsasser[9] who used the Laue criteria for x-ray scattering. Quantitative interpretation was impossible since the percentage of the neutrons which was scattered elastically was unknown. Shortly afterwards Mitchell and Powers[10] observed the Bragg reflection of neutrons from MgO single crystals and established conclusively the diffraction of

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[1] Whitaker, Beyer and Dunning, Phys. Rev. **54**, 771 (1938).

[2] M. D. Whitaker and H. G. Beyer, Phys. Rev. **55**, 1124 (1939).

[3] M. D. Whitaker and H. G. Beyer, Phys. Rev. **55**, 1101 (1939).

[4] Halpern, Hamermesh and Johnson, Phys. Rev. **55**, 1125 (1939).

[5] G. C. Wick, Physik. Zeits. **38**, 403 (1937); **38**, 689 (1937).

[6] O. Halpern and M. H. Johnson, Phys. Rev. **55**, 898 (1939).

[7] I. Pomerantschuk, Physik. Zeits. Sowjetunion **13**, 65 (1938).

[8] P. Preiswerk and H. von Halban, Comptes rendus **203**, 73 (1938).

[9] W. M. Elsasser, Comptes rendus **202**, 1029 (1936).

[10] D. P. Mitchell and P. N. Powers, Phys. Rev. **50**, 486 (1936).

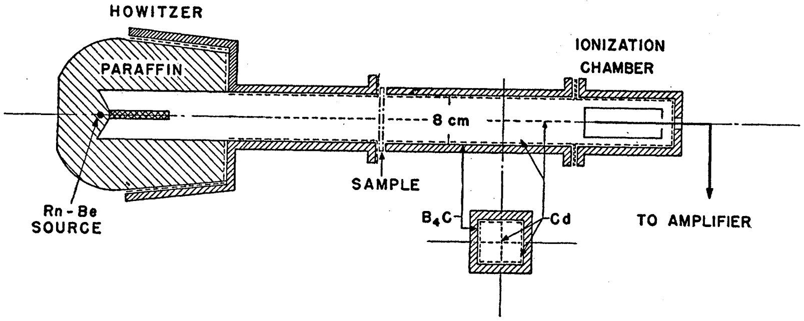

FIG. 1. Collimation and shielding for producing thermal neutron beam.

slow neutrons. A later experiment by Preiswerk[11] on Al crystals was consistent with the work of Mitchell and Powers.

In this paper we are reporting measurements of C neutron interactions as measured by total cross sections. This method as contrasted with the methods employed previously yields quantitative results. Our measurements are divided into three parts: (1) Measurement of the cross section of the same element in different crystalline states. (2) Measurement of the cross section of alloys. (3) Measurement of the cross section of chemical compounds.

## EXPERIMENTAL ARRANGEMENT AND PROCEDURE

### A. Neutron beam and detecting apparatus

The neutron sources used in this work were radon-beryllium bulbs initially containing about 600 millicuries of radon. These source bulbs were placed in a paraffin "howitzer" the details of which are shown in Fig. 1. The use of such howitzers for the production of collimated neutron beams has been described in detail by Fink.[12] From the howitzer opening, the neutrons passed down a rectangular channel 8 cm×8 cm, the walls of which were filled with $B_4C$ to a thickness of about 1 cm as shown in the diagram. This channel was lined with cadmium and was divided into 4 rectangular tubes 4×4 cm by cadmium. It was possible to use only one of these tubes in cases where only small samples were available. This arrangement assured us that any C neutron emerging from one of these tubes has passed down the tube without striking the wall since cadmium has such a large capture cross section for these neutrons. There was an opening at the center of this collimating system in which the samples were placed. Since the distance from the source to the central opening in the beam was 40 cm, the neutrons striking the scatterer when it was placed in the middle position were well collimated. The collimating tube between the scatterer and the detector assured that any neutrons scattered through angles greater than four degrees would fail to reach the detector. It was necessary to modify the neutron beam in order to use the small single crystals of Ni, Ni-Fe, Fe, and $SiO_2$. This was done by putting a diaphragm of $B_4C$ and Cd in the beam in such a way that each sample would just cover the opening in the diaphragm.

The detectors were ionization chambers either lined with boron carbide or filled with boron trifluoride. These chambers were used with a linear amplifier and a scale-of-two counter.

### B. Preparation of samples

Where possible the samples used were of such a size that they covered the entire 8×8-cm neutron beam and were of such a thickness that 30 percent to 40 percent of the slow neutrons traversing them were removed from the detected beam. With samples of this thickness double scattering was small. The chemical compounds and other materials which were in powdered form were placed in sample containers made of copper sheet 0.020 cm thick which were sealed gas tight. Experimental corrections were made for the copper sheet in these cases.

Materials of carefully determined purity were used. Reagent grade chemicals were procured and in most cases these were re-analyzed. Water was an "impurity" which had to be controlled very carefully. The very high cross section for C

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[11] P. Preiswerk, Helv. Phys. Acta **10**, 400 (1937).
[12] G. A. Fink, Phys. Rev. **50**, 738 (1936).

neutrons and low atomic weight of $H_2O$ made it necessary to use carefully dried samples, since in most of the work we were interested in differences between measurements on two or more samples. In some cases a water content of $\frac{1}{10}$ of one percent by weight in one of these samples would have contributed more to the apparent cross section of the material than the difference being measured.

We are much indebted to Mr. Gabor Levy for developing a drying technique which proved to be highly satisfactory. The essential details of the drying apparatus are shown in Fig. 2. The apparatus consists of a large glass tube *A* which holds the material. This tube is of Pyrex glass of 5 cm diameter and is 55 cm long. A ground joint allows access to the tube through a full diameter opening. This tube is heated by a water bath whose temperature is controlled electrically. The large tube is connected to a Hyvac pump and a large bottle containing $P_2O_5$, through either a glass tube *B* or a detector tube *C*. These two tubes are interchangeable.

The usual procedure was to place the material in the large tube *A* and close the apparatus using the tube *B* to connect it to the pump and $P_2O_5$ bottle. The apparatus was evacuated and the large tube heated to 70° or 80°C. The apparatus was pumped continuously and maintained at the above temperature until there was no longer any visible moisture on the colder parts of the system. After the distillation ceased to be visible the tube *B* was replaced by the absorber tube *C*. This is a tube for the quantitative absorption of water as used in the microanalytical methods of Pregl. It is filled with "anhydrone" ($Mg\,ClO_4$) which is a quantitative absorber of water. During the last stages of a drying operation this tube was placed in position for 15 minutes periodically and a graph made of water absorbed in 15 minutes as a function of time. It was found in all cases that the water absorbed by the anhydrone tube approached zero and the drying operation was considered complete when the increase in weight of the tube was no longer measurable. The sample material was usually left in the dryer for 1 to 3 days.

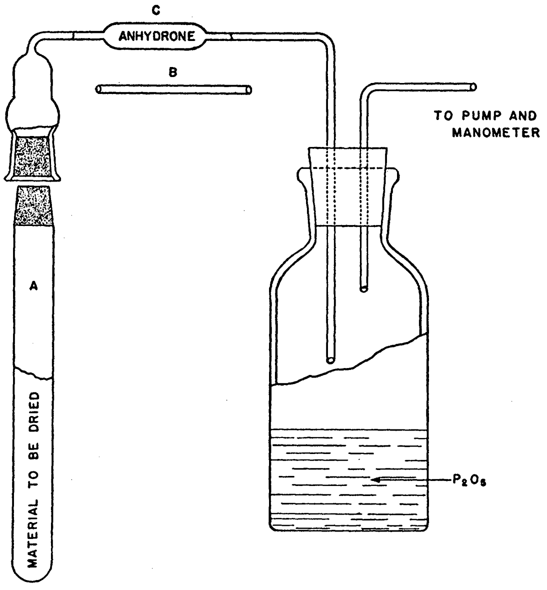

FIG. 2. Drying apparatus.

To test the drying efficiency of the apparatus the following experiment was performed. Some very fine powdered $SiO_2$ (sand) was heated to 600°C and was found to lose about 1 percent in weight. Continued heating at 600°C resulted in no further loss in weight so it was considered to be dry. Distilled water was added to this dry material until it was 30 percent water by weight. This sample was placed in the apparatus and its loss of weight was followed. At the end of 3 hours about 2 percent of the water remained in the material while at the end of 6 hours about 0.1 percent remained. Further tests showed that the anhydrone tube absorbs all the moisture removed from the sample if it is placed in the system after visible evidences of evaporation have disappeared.

The chemical compounds used were usually found to contain from a fraction of 1 percent up to 2 percent moisture before drying. It is believed that this drying technique removed moisture, other than water of crystallization, until less than 0.01 percent by weight remained. This method has been used to dry materials satisfactorily which give up or take on oxygen at temperatures less than 100°C.

## C. Cycle of operation

All the data on crystals, alloys, compounds, and elements were taken in the form of cycles containing four observations. These observations were: (1) The number of disintegrations, $N_1$, produced in the boron ionization chamber in 20 minutes by the neutrons traversing the collimating tubes and passing through no material. (2) The number of disintegrations, $N_2$, produced in the same time interval with 0.8 g/cm$^2$ of cadmium in the beam. (3) The number of counts, $N_3$, produced with the scattering material in the beam. (4) The number of counts, $N_4$, produced with both scattering material and cadmium in the beam.

The ratio, $N_3-N_4/N_1-N_2$ is called the transmission, $P$, of the material. The cross section was calculated from the transmission from the formula $P=e^{-\sigma NX}$ where $\sigma$ is the cross section (total), $N$ the number of nuclei per cm$^3$, and $X$ the thickness in cm. In cases where we were interested in comparing the transmission of two given materials, the two were used in the same cycle, the cycle being extended to 6 observations to include transmission of the added sample both with and without Cd. The apparatus counted from 100 to 500 neutrons per minute with no scattering material in place. The number of counts per minute varied linearly with the strength of the source, indicating that the scale-of-two counter was not failing to resolve an appreciable number of pulses at the higher counting rates. As an added precaution, however, the scattering samples were made to have about the same transmission and were used with both strong and weak sources and the results compared. Any correction for neutrons which were scattered by the material under observation and were still detected contributed less than 1 percent to the computed value of the measured cross sections.

## RESULTS

### A. Measurements of cross sections of the same elements or compounds in different crystal states

Measurements were made on quartz, iron, and nickel, in both the polycrystalline and single crystal states. Where possible, measurements were made on two thicknesses of single crystal. The number of g/cm$^2$ is given where this is the case. In the case of quartz, measurements were also made to determine the cross section of the $SiO_2$ molecule in the fused state. For this last measurement fused quartz cylinders from a Fabry-Perot interferometer were used. The iron used was not in the form of a single crystal but was composed of several large crystals which extended from one face to the other so that each part of the neutron beam traversed a single crystal. The results are given in Table I below in the form of cross sections for individual atoms or molecules for each crystalline state.

TABLE I. *Cross sections of elements and compounds in different crystal states* $\times 10^{24}$ *cm*$^{-2}$.

| | AMORPHOUS | POLYCRYSTALLINE | SINGLE CRYSTAL | G/CM$^2$ |
|---|---|---|---|---|
| Fe | | 12.0±0.2 | 7.0±1.0 | 1.6 |
| | | | 6.1±1.0 | 8.8 |
| Ni | | 19.8±0.5 | 14.1±1.2 | 4.4 |
| $SiO_2$ | 8.0±1.0 | 8.8±1.0 | 4.5±0.6 | 3.7 |
| | (fused) | (sand) | 4.1±0.6 | 1.3 |

### B. Measurements of cross sections of alloys

In Table II are given similar results of measurements on the cross sections of alloys. The Permalloy samples were the usual iron-nickel alloys which are heat treated to enhance their initial permeabilities. Here the measurements on single crystals and on polycrystalline material were not made on alloys of exactly the same composition, as is indicated. Measurements were also made on a sample of copper zinc alloy of 40 atomic percent copper but there was no notice-

TABLE II. *Cross sections of alloys* $\times 10^{24}$ *cm*$^{-2}$.

| | ADDITIVE* | POLYCRYSTALLINE | SINGLE CRYSTAL |
|---|---|---|---|
| Permalloy (78) | 18.2 | 12.5±0.7 | |
| Permalloy (73) | 17.8 | | 10.2±1.3 |
| Permalloy (68) | 17.4 | | 10.2±1.3 |
| Permalloy (45) | 15.6 | 16.0±0.8 | |

* Additive value computed from cross sections of polycrystalline iron and nickel.

TABLE III. *Cross sections of compounds* $\times 10^{24}$ *cm*$^{-2}$.

| COMPOUND | MEASURED CROSS SECTION | ADDITIVE CROSS SECTION |
|---|---|---|
| MnO | 19.9 | 17.2 |
| MnS | 19.1 | 15.1 |
| $MnO_2$ | 25.2 | 21.3 |
| $MnSO_4$ | 33.6 | 31.5 |
| $Fe_2O_3$ | 39.2 | 36.3 |
| CuO | 16.2 | 14.6 |
| CuS | 16.5 | 12.5 |
| $Cu_2O$ | 29.5 | 25.1 |
| $Cu_2S$ | 27.7 | 23.0 |
| ZnO | 7.7 | 8.6 |
| ZnS | 9.7 | 6.5 |
| NiO | 22.3 | 23.9 |
| HDO (See text) | 54 | 54.5 |
| $D_2O$ | 16 | — |

able deviation from the result to be expected from the copper and zinc content.

### C. Measurements on compounds and elements

In Table III is given a list of the cross sections per molecule of certain chemical compounds which have been measured. The last column of this table gives the sum of the atomic cross sections of the constituent elements. These values are taken from Table IV. The "compound" listed as HDO was obtained by mixing equal parts of $H_2O$ and $D_2O$. The subsequent measurements of Kimura[13] on physical and chemical mixtures of Hg and S, are in good qualitative agreement with our results.

In most cases about $10^5$ neutrons were counted in determining each cross section. The above values should represent the actual cross section to within ±3 percent.

In studying the effect of crystalline and chemical form on the cross section of various alloys and compounds it was necessary to measure the total cross sections of the elements involved in some well-known form under the same conditions. In Table IV is given a list of the cross sections of each of the elements occurring in the crystals, alloys, and compounds mentioned above.

TABLE IV. *Cross sections of elements* $\times 10^{24}$ *cm*$^{-2}$.

| ELEMENT | FORM | MEASURED CROSS SECTION |
|---|---|---|
| C | (Graphite) | 4.87±0.1 |
| Cu | Rolled-plate | 10.5 ±0.4 |
| Fe | Armco | 12.0 ±0.2 |
| Mn | Metallic grains | 13.1 ±0.6 |
| Ni | Rolled sheet | 19.8 ±0.5 |
| O | Gas[1] | 4.1 |
| S | Powder | 2.0 ±0.6 |
| Zn | Metallic grains | 4.5 ±0.5 |

[1] H. Carroll and J. R. Dunning, Phys. Rev. **54**, 541 (1938).

## DISCUSSION OF RESULTS

The properties of C neutrons have been investigated by various methods. The velocity distribution of the C neutrons has been measured directly with a cadmium velocity selector.[14] From these measurements it was concluded that most of the C neutrons have energies corresponding to thermal velocities with a most probable velocity at 300°K near the value $v=(2KT/M)^{\frac{1}{2}}$. This implies about *1.7* angstrom units as the most probable wave-length for the distribution. The albedo measurements of Amaldi and Fermi[15] showed that C neutrons can traverse many free paths in a hydrogenous material while belonging to that group thus indicating that the cadmium absorption band includes neutrons of thermal energy.

In the case of single crystals only small bands of the neutron spectrum can satisfy conditions analogous to the Laue conditions for x-rays, so that for the distribution as a whole we should expect the single crystals to be quite transparent. In polycrystalline material on the other hand various orientations of the crystallites make it possible for the spectrum as a whole to be scattered.

From Table I we see that in every case single crystals of a given material are much more transparent than the same material in a polycrystalline form. Our results showed no marked dependence upon the thickness so that we may conclude that only a small part of the spectrum is scattered selectively by a single crystal orientation.

In the case of Fe, Ni, and $SiO_2$ measurements, the only variable was the size of the crystallites, and therefore the effects observed must be attributed to change in crystallite size. The results for the Fe-Ni alloys are complicated by the fact that the same conditions which favor single crystal formation may increase the order in the alloy.

Had these experiments been performed with monochromatic neutrons of various wave-lengths the crystals would probably have shown an even greater transparency for the various wave-lengths, provided they are not too short, with the exception of a few selectively scattered bands. For the distribution actually used some of the C neutrons are scattered because their wave-length is so short that they do not produce these interference effects.

The results on alloys are more complicated, since here a change of transmission of 2 metals when mixed together in an alloy may result

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[13] M. Kimura, Proc. Imp. Acad., Tokyo **15**, 214 (1939).

[14] Dunning, Pegram, Fink, Mitchell and Segrè, Phys. Rev. **48**, 704 (1935).

[15] E. Amaldi and E. Fermi, Phys. Rev. **50**, 899 (1936).

because: (1) A change in the crystal lattice and crystalline grain size occurs, or (2) Each constituent element may now have unlike neighbors depending on the state of order, if any, of the crystallites.

If the transmission increase in Permalloy 78 results from (2), the sensitivity of the neutron transmission to order in the alloy may have resulted from large phase shift differences between waves scattered by Fe and Ni. Subsequent experiments[16] show that the transmission depends on the order in the alloy. In these experiments two samples were prepared by appropriate heat treatment so that (1) is the same for both the samples while (2) is different for the two samples.

The results on alloys indicate that the resultant effects in chemical compounds are not necessarily of a chemical nature, as in all chemical changes in the solid state there is an attendant change in crystalline state.

The results on compounds are the most difficult to interpret because one compares the cross sections of compounds in polycrystalline states with the cross sections of the elements which compose them. The constituent elements sometimes were polycrystalline, liquid, or even gaseous. Here the effect may be physical or chemical, and is complicated by the possibility of large phase changes in the scattered wave from the different constituent nuclei.

If the results on Ni$-$Fe 78 are interpreted as evidence for large phase shift differences between the waves scattered by the Ni and Fe nucleus, one should expect either $Fe_2O_3$ or NiO to be more transparent than a physical mixture of the pure elements and this was observed to be true for NiO. This is based on the assumption that phase shift differences close to $\pi$ are in general more effective in modifying interference phenomena, than changes in spacing.

It is interesting that the measurements on a 50 percent $D_2O$ and 50 percent $H_2O$ mixture showed no deviation from additivity.

Our experiments show that general interference phenomena will complicate the interpretation of any change in scattering which results from a change in the molecular composition of the material. The scattering of neutrons from magnetic materials is a rather interesting case of the above.[1]

In general, results from C neutrons are difficult to evaluate. Such phenomena as the change of scattering cross section of a given material between the C group and groups of higher energy may be attributed to interference effects rather than a change in the nuclear cross section with change in energy of the neutron. Any deductions from C neutron measurements which depend upon the assumption of spherically symmetric distribution of the scattered neutrons will not be rigorously valid.

Measurements using neutrons having energies well above that of the C neutrons should be free from these interference effects.[17, 18] If this can be checked by experiments similar to our measurements, cross sections measured with these neutrons, in contrast with C neutrons, can be interpreted as the cross section per isolated atom, at least in the case of heavy elements.

We take pleasure in acknowledging the courtesy of Dr. R. M. Bozorth, Mr. P. P. Cioffi, and Mr. O. L. Boothby of the Bell Telephone Laboratories who have furnished the single crystals of Ni and Ni-Fe alloys which were used. We wish to thank Dean G. B. Pegram and Professor J. R. Dunning for their coöperation and encouragement during the progress of the work.

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[16] Nix, Beyer and Dunning, Phys. Rev. **57**, 566 (1940).

[17] H. B. Hanstein and J. R. Dunning, Phys. Rev. **57**, 565 (1940).

[18] M. D. Whitaker, W. C. Bright and E. J. Murphy, Phys. Rev. **57**, 551 (1940).