Thermal Neutron Scattering Studies in Metals

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The interaction of slow or C neutrons can be modified by changes in the physical parameters of the material which the neutrons are traversing. Measurements of the effect of grain size on *total* C neutron cross section were made on randomly-oriented poly-crystalline iron and copper. In the case of copper it was found that the total cross section decreased in an exponential manner with increase in grain size. For iron it was simply established that the total cross section was substantially smaller for the larger grain size.

THE work of Nix, Beyer, and Dunning[1] has shown that the presence of order in binary iron-nickel alloys, in which the grain size was maintained constant and free of preferred orientation, decreased the cross section below that obtained from random alloys: the greater the degree of order, the lower the total cross section obtained. The establishment of such an effect[1,2] illustrates that thermal neutrons may be used as a tool to study the structure of the solid state. In order to render this tool more useful for our systematic studies of order-disorder transformations in alloys, it was necessary to establish the influence of other physical parameters on the neutron scattering. As our first problem in this series of studies, we selected the effect of grain size. Grain growth frequently takes place in the thermal treatments necessary to produce order-disorder transformation. As metals we selected copper as a non-ferromagnetic face-centered cubic metal free from polymorphic transformations, and iron as a ferromagnetic body-centered cubic metal subject to polymorphic transformations.

EXPERIMENTAL ARRANGEMENT AND PROCEDURE

The neutron source was a radium-beryllium bulb containing 200.5 mg of radium and 8.0 mg of beryllium. Figure 1 shows the detailed arrangement of the "howitzer," scatterer, ionization chamber, amplifier, and counting circuit. Thermal neutrons from the "howitzer" passed down a cylindrical collimating tube 10 cm in diameter. The interior wall of the tube was lined with cadmium. The collimating tube contained a 0.635-cm hollow outer shell which was filled with B_4C in order to reduce the number of neutrons escaping into the room. The length of collimating tube from source to test sample was 27 cm. The collimating tube was constructed in two parts permitting samples to be placed in the neutron beam at the center of the system. The collimating tube between sample and detector was 26 cm long which prevented the C neutrons scattered at an angle of greater than 8 degrees from reaching the detector.

The neutron detector was a BF_3 ionization chamber connected to a linear amplifier and scaling system recorder located in a distant room in the laboratory building.

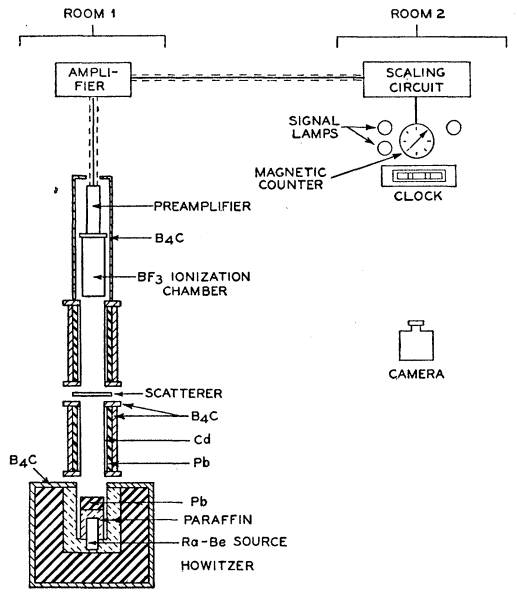

FIG. 1. Experimental arrangement.

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[1] F. C. Nix, H. G. Beyer, and J. R. Dunning, Phys. Rev. **58**, 1031 (1940).

[2] H. G. Beyer and M. D. Whitaker, Phys. Rev. **57**, 976 (1940).

The samples to be tested were automatically introduced into and removed from the neutron beam by electric motors. The sequence of operation of the motors was controlled by a timing circuit which provided for the following neutron beam conditions:

1. Open beam.
2. Cadmium slab in beam.
3. Sample and cadmium slab in beam.
4. Sample in beam.

When comparing the relative transmission of two samples we use the 1, 2, and 4 conditions with the cadmium replaced by the second sample. The 1 condition permitted us to keep a frequent check on amplifier and scaling circuit conditions. The timing circuit actuated the motors to operate at intervals varying from 3 to 60 minutes. All measurements in this research were carried out at 80-minute cycles, which allowed 20 minutes for each part of the cycle.

The data were recorded automatically by photographing simultaneously the counter, an electric clock which could be read to 0.1 second, and signal lamps. The use of the clock permitted the duration of each part of the cycle to be accurately measured. The combination of the three signal lamps gave positive information as to which of the above-listed beam conditions prevailed at any instant. A 16-mm motion picture camera, provided with a feature allowing the exposure of one frame at a time, was used to photograph the data, which included clock, signal lamps, and counter. Exposures were taken at intervals of 45 seconds.

The thickness of the scattering sample was such that it removed approximately 40 percent of neutrons from the "open" beam.

PREPARATION OF SAMPLES

The copper samples were prepared from an ingot of very pure deoxidized high conductivity copper by hot-working plates of the material at a temperature of 1000°C to 50 percent of their initial thickness of 3.05 cm, cold working to a thickness of 0.76 cm, and milling to a final thickness of 0.63 cm. The samples were plates $12.7\times12.7\times0.63$ cm.

The various grain sizes were obtained by annealing the cold worked material. The annealed iron sample was obtained by vacuum annealing at 1000°C for 30 minutes. Seven copper samples were prepared by vacuum annealing for 30 minutes at each of the following temperatures, 450°C, 500°C, 550°C, 650°C, 750°C, 800°C, 850°C, while an eighth sample was annealed at 1000°C for one hour. Cold worked samples of iron and copper were also retained.

The grain sizes of all scattering samples were obtained from photomicrographs of the polished and etched specimen. The qualitative orientation of the crystallites in each sample was established from x-ray diffraction patterns taken at grazing angles. The x-ray and microscopic examinations were made on the portion of the specimen exposed to the neutron beam.

METHOD OF MEASUREMENTS

a. Relative Neutron Transmission Measurements

The relative neutron transmission measurements simply consist in obtaining the transmission of two identically-shaped plates in the neutron beam. The number of counts per minute, N_a, registered in the ionization chamber with annealed sample as scatterer, is compared with the number obtained with cold worked sample as scatterer, N_b.

The transmission of the annealed sample relative to the cold worked sample, T_{ab}, is expressed by,

$$T_{ab}=\frac{N_a-N_b}{N_b}\times100. \qquad (1)$$

The final value of relative transmission was taken as the average of 36 cycles.

b. Measurement of Total Cross Section of Scatterer

The total cross section is obtained from the following four measurements:

1. The number of disintegrations produced in the BF_3 ionization chamber per minute, N_1, by the open beam with no scatterer present.
2. The number of disintegrations per minute, N_2, with 0.45 g/cm² of cadmium in the neutron beam.
3. The number of disintegrations per minute, N_3, with both cadmium and the scatterer whose cross section is to be determined in the beam.
4. The number of disintegrations per minute, N_4, with the scatterer alone in the neutron beam.

The cadmium in parts 2 and 3 of the cycles serves to prevent any thermal neutrons from reaching the ionization chamber.

The transmission, T, of the scatterer for C neutrons can then be determined from the following equation,

$$T=(N_4-N_3)/(N_1-N_2). \quad (2)$$

The total cross section of the scatterer can be calculated from the equation,

$$T=e^{-\sigma nt} \quad (3)$$

where σ is the total cross section in sq. cm; n, the number of nuclei per cubic cm of scatterer; and t, the thickness of the scatterer in cm. The number of nuclei per cm^3 can be obtained from the formula,

$$n=d\times 6.02\times 10^{23}/\text{Atomic weight}, \quad (4)$$

where 6.02×10^{23} is Avogadro's number and d is the density of the scatterer.

The cross-section values given in Tables I and II were taken from the average of 18 determinations. The number of neutrons recorded in the ionization chamber in the open beam condition was approximately 1000 per minute.

DISCUSSION OF RESULTS

The percent changes in transmission, referred to the cold worked sample, for measurements on iron and copper samples are given in Table I. These values are obtained from Eq. (1). They show the annealed samples to possess the larger transmission; with random orientation of crystallites, the larger the grain size the greater the transmission.

The total cross-section values, σ, obtained on cold worked and annealed samples are given in Table II. These values were obtained by the use of the Eq. (3) from transmission values obtained from Eq. (2). In Table II is also included information on preparation of samples, values of grain sizes determined microscopically, and comments on the orientation of the crystallites.

TABLE I. Relative thermal neutron cross sections.

Material	Preparation of sample	Grain size in millimeters	Orientation	Percent change in transmission referred to cold worked sample
Fe	Cold worked	0.077	Random	+7±2.0
Fe	Annealed ½ hr. 1000°C	0.127	Random	
Cu	Cold worked	0.005	Random	+9±2.0
Cu	Annealed ½ hr. 1000°C	0.081	Random	

TABLE II. Thermal neutron cross section as a function of grain size $\times 10^{24}$ cm^{-2}.

Material	Preparation of sample	Grain size in millimeters	Orientation	Total cross-section σ, $\times 10^{24}$ cm^{-2}
Fe	Cold worked	0.077	Random	13.06±0.4
Fe	Annealed ½ hr. 1000°C	0.127	Random	11.39±0.3
Cu	Cold worked	0.005	Random	10.84±0.3
Cu	Annealed ½ hr. 450°C	0.017	Random	10.16±0.4
Cu	Annealed ½ hr. 500°C	0.015	Random	10.13±0.5
Cu	Annealed ½ hr. 550°C	0.021	Random	9.96±0.4
Cu	Annealed ½ hr. 650°C	0.036	Random	9.62±0.3
Cu	Annealed ½ hr. 750°C	0.373	Random	7.76±0.2
Cu	Annealed ½ hr. 800°C	0.567	Random	7.58±0.3
Cu	Annealed ½ hr. 850°C	0.640	Random	7.37±0.4
Cu	Annealed 1 hr. 1000°C	2.40	Random	7.04±0.3

A plot of total C neutron cross section, σ, *versus* grain size for copper is given in Fig. 2. The *larger* the grain size, the smaller the *total cross section.* It is to be noted from Table II that all samples were free from any detectable preferred orientation of the crystallites. These results clearly show that the total C neutron cross sections of solid material cannot be looked upon as physical constants of the elements or compounds in question but are largely determined by the diffraction effects within the body of the scatterer. With experimental arrangement given in Fig. 1 each Bragg reflection tends to remove neutrons from the well collimated beam, causing them to be deflected to the cadmium-lined collimator tube wall and consequently removing them from the beam. For *completely randomly* oriented crystallites, the greater the number of crystallites intercepting the beam the greater the decrease in the transparency of the sample. The data in Tables I and II and the graphical illustration of the copper data in Fig. 2 clearly show the increase in transparency with the consequent decrease in total cross section

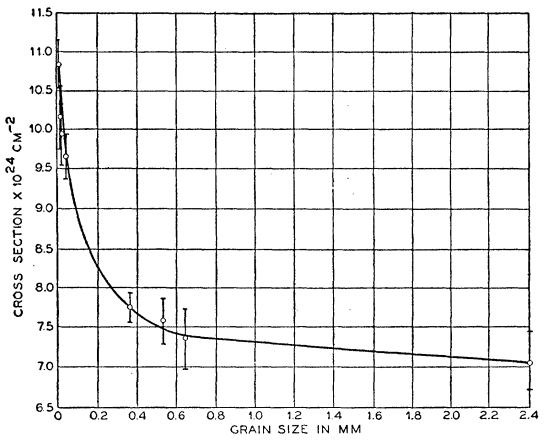

FIG. 2. Grain size *vs.* cross section $\times 10^{24}$ cm^{-2} for copper.

with increasing grain size. With random orientation, the larger the grain size, the fewer grains are available for Bragg reflection.

These experiments clearly show that the thermal neutron cross-section values given in literature lose much of their supposed quantitative significance and should be regarded as purely qualitative in nature. As an illustration, the values for copper depend to a large extent on the grain size and would be further complicated by the presence of preferred orientation. In the case of alloys there is the additional complication of atomic distribution in solid solutions and spatial distribution of phases for heterogenous binary alloys. In the work of Nix, Beyer, and Dunning[1] on iron-nickel alloys in the region around 75 atomic percent nickel, it was shown that the presence of ordering tends to increase the transparency to thermal neutrons. In this work the grain size was maintained constant while varying the degree of order, and there was demonstrated an increase of as much as 8 percent in transparency on increasing the degree of order.

As stated in the introduction, this investigation was undertaken with the object of exploring the possible use of neutrons as a tool for investigating the solid state. In order to evaluate properly data obtained at elevated temperatures on order-disorder or other solid-state transformations, it was found to be necessary to establish the effect of parameters such as grain size and change in orientation of crystallites on neutron cross-section values.

It appears that the use of thermal neutrons with wave-lengths comparable to the interatomic spacings in crystals will be a very useful method for studying the structure of the solid state. Accurate quantitative information permitting unequivocal interpretation of changes in the solid state must await the development of intense sources of monochromatic neutron beams; however, much valuable information can be obtained from the use of a relatively weak radium-beryllium source of wide wave-length bands such as were used in the present investigation.