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A New Method for Determining the Relative Phase with Which Slow Neutrons Are Scattered by Nuclei*

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To determine the relative phase with which neutrons are scattered by an element A, cross-section measurements are obtained for A, for a reference element B, and for a substitutional solid solution of A and B. The experimental value of the effective coherent scattering cross section, σ_{coh}', for the solid solution, when compared with the values calculated from the data for pure A and B, shows whether the two constituent elements scatter neutrons in the same or in opposite phases. Measurements made with the Columbia neutron velocity spectrometer show that copper and nickel scatter with the same phase, and manganese and nickel with opposite phases, in agreement with other methods of phase determination. The following more accurate values of the free scattering cross sections, σ_f, and new values of the effective coherent scattering cross sections, σ_{coh}', were obtained: copper, $\sigma_f=8.2$ barns, $\sigma_{coh}'=6.6$ barns; nickel, $\sigma_f=17.4$ barns, $\sigma_{coh}'=13.9$ barns; manganese, $\sigma_f=1.80$ barns. $\sigma_{coh}'=\sim 1.9$ barns.

INTRODUCTION

In the course of the systematic investigation of the resonance levels of the elements being carried out with the Columbia neutron velocity spectrometer,[1,2] the free scattering cross sections and the $1/v$ capture slopes of the elements are determined with considerable precision. With relatively little additional work, as will be shown in this paper, it is possible to determine the coherent scattering cross section and the relative phase with which an element scatters neutrons. This new method does not require high resolution, and hence there is no need for the high neutron intensity of a reactor. The data are easily analyzed and the results are obtained directly.

Fermi and Marshall[3] have reported three methods for phase determination: (a) Intensity measurements of different orders of Bragg reflections from the same plane of a crystal and comparison with theoretical values; (b) measurement of the limiting angle for total reflection of neutrons from mirrors; (c) comparison of the observed total cross section of gas molecules at long neutron wave-lengths with values calculated from classical interference theory. The second method is the only one which gives an absolute determination of phase, for only nuclei with positive scattering amplitudes, using the Fermi sign convention, give total reflection. The first two methods require a high neutron flux in order to obtain good resolution in a reasonable period of time. The third method is not a precision experiment and does not require high neutron intensities, but it is limited in its application to gases.

Wollan and Shull[4] made phase determinations by observing the coherent Bragg scattering by different crystal planes, and comparing with theoretical values. Fermi, Sturm, and Sachs[5] and Winsberg, Meneghetti, and Sidhu,[6] have determined phase of scattering by making high resolution transmission measurements of microcrystals, and comparing crystal diffraction peaks of the transmission curve, after identification of the peaks with particular crystal planes, with theoretical values.

THEORY

The theory of the phase of scattering of slow neutrons has been treated in earlier papers.[3,4,7,8] Only the results

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[1] Havens, Rainwater, Wu, and Dunning, Phys. Rev. **73**, 963 (1948).

[2] Rainwater, Havens, Dunning, and Wu, Phys. Rev. **73**, 733 (1948).

[3] E. Fermi and L. Marshall, Phys. Rev. **71**, 666 (1947).

[4] E. O. Wollan and C. G. Shull, Phys. Rev. **73**, 830 (1948).

[5] Fermi, Sturm, and Sachs, Phys. Rev. **71**, 589 (1947).

[6] Winsberg, Meneghetti, and Sidhu, Phys. Rev. **75**, 975 (1949).

[7] Halpern, Hamermesh, and Johnson, Phys. Rev. **59**, 981 (1941); also O. Halpern and M. H. Johnson, Phys. Rev. **55**, 898 (1939).

[8] Feshbach, Peaslee, and Weisskopf, Phys. Rev. **71**, 145 (1947).

of this theory, and the definitions used in this paper, need be mentioned here. For slow neutron scattering by nuclei, only zero angular momentum or spherically symmetric scattering need be considered. If η is the phase shift relative to the incident neutron wave of the spherically symmetric part of the total wave at large distances, then the free scattering cross section, σ_f, is given by

$$\sigma_f=4\pi(-\lambda\bar{\ }\sin\eta)^2, \tag{1}$$

where $\lambda\bar{\ }=\lambda/2\pi=1/k$ and λ is the neutron wave-length. For scattering by a nucleus of spin zero, $(-\lambda\bar{\ }\sin\eta)$ has a unique value; this quantity is called the free scattering amplitude or scattering length and is denoted by a. For $|\sin\eta|\ll 1$, a is just the value of the radial coordinate r for the (extrapolated) node of the exterior wave function. From general theoretical considerations, the free scattering length is expected to be positive, except in a small energy region on the low side of a resonance, where it is negative. If two nuclei scatter neutrons with the same phase, then the sign of a is the same for both; if they scatter with opposite phases, then a is positive for one nucleus and negative for the other.

If the spin i of the nucleus is not zero, then compound nuclear states of spin $(i+\frac{1}{2})$ and $(i-\frac{1}{2})$ can be formed with relative statistical weights $g_a=(i+1)/(2i+1)$ and $g_b=i/(2i+1)$, respectively. If the corresponding free scattering lengths are a_a and a_b, then the total free scattering cross section for the isotope is

$$\sigma_f=4\pi[g_a a_a^2+g_b a_b^2]. \tag{2}$$

This is the experimentally observed cross section using neutrons of energy >1 ev. At lower energies the binding of the atoms is important and the scattering amplitudes for an infinitely bound atom are increased by the factor $(A+1)/A$, where A is the atomic weight of the nucleus. The bound scattering cross section, σ_b, for a single atom is then

$$\sigma_b=4\pi[g_a f_a^2+g_b f_b^2], \tag{3}$$

where f_a and f_b are the bound scattering amplitudes for the separate spin states.

In a crystalline material containing many atoms, we must take into account that part of the scattering by a single nucleus is coherent relative to scattering by the other atoms, and part is incoherent. The observed scattering at any given energy below about 1 ev is very sensitive to the crystal lattice parameters and to the neutron wave-length on account of interference between the coherent part of the scattering from different atoms. We can define the coherent scattering cross section, σ_{coh}, as that part of σ_b which can be modified by interference effects. Then σ_{coh} is given by

$$\sigma_{coh}=4\pi[g_a f_a+g_b f_b]^2. \tag{4}$$

If measurements are made on an element with several isotopes of relative abundance p_j present, the above averages must also be taken over the isotopes. Then

$$\sigma_f=4\pi\sum_j p_j[g_{aj}a_{aj}^2+g_{bj}a_{bj}^2], \tag{5}$$

$$\sigma_b=4\pi\sum_j p_j[g_{aj}f_{aj}^2+g_{bj}f_{bj}^2], \tag{6}$$

and

$$\sigma_{coh}=4\pi\{\sum_j p_j[g_{aj}f_{aj}+g_{bj}f_{bj}]\}^2. \tag{7}$$

From Eq. (7) we can define the effective bound scattering amplitude, f_{k-eff}, for an element k consisting of more than one isotope by

$$f_{k-eff}=\sum_j p_j[g_{aj}f_{aj}+g_{bj}f_{bj}]. \tag{8}$$

It is the sign of this effective scattering amplitude which is determined by the experiments. The incoherent scattering cross section, σ_{inc}, of an element due to spin and isotope effects is given by

$$\sigma_{inc}=(\sigma_b-\sigma_{coh}). \tag{9}$$

If solid solutions are formed, of two metals for example, there are two cases to consider. If the system is completely disordered with respect to the relative positions of the atoms of the two elements in the lattice, then we may still use Eqs. (5)–(7), but treat p_j as the relative abundance of the jth isotope in the mixture rather than in the element. If the coherent scattering amplitudes are quite different (or of opposite sign) for

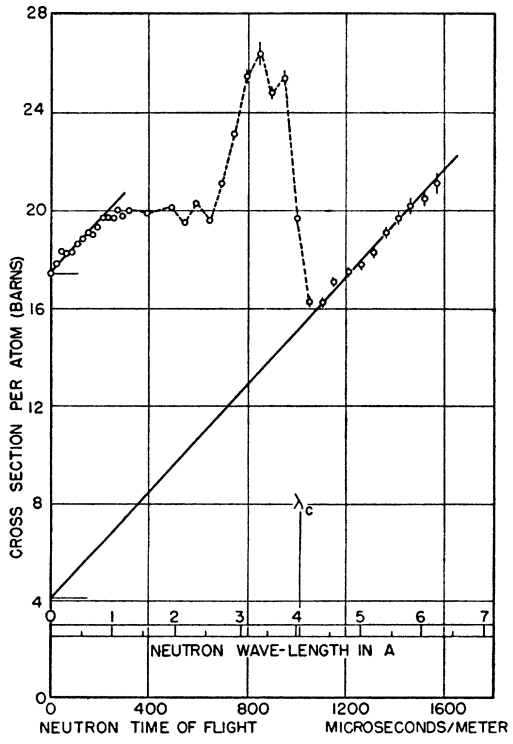

FIG. 1. The slow neutron cross section of nickel as measured with a 9.69-g/cm² sample. The $1/v$ line for the short wave-length region is $\sigma=(17.4+0.79E^{-\frac{1}{2}})$, and the $1/v$ line for data beyond the cut-off for Bragg reflections, λ_c, is $\sigma=(4.1+0.79E^{-\frac{1}{2}})$.

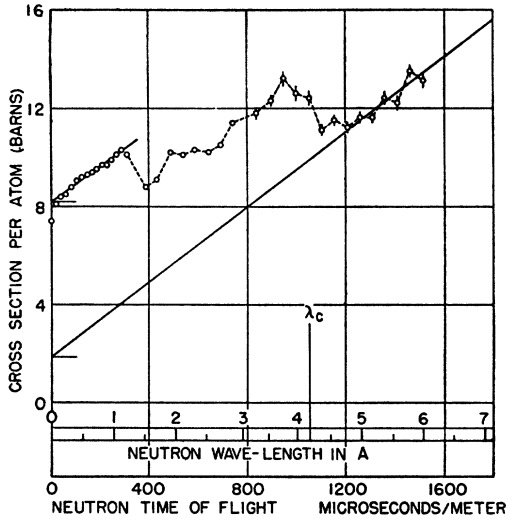

FIG. 2. The slow neutron cross section of copper as measured with a 19.19-g/cm² sample. The $1/v$ line for the short wave-length region is $\sigma=(8.2+0.54E^{-\frac{1}{2}})$, and the $1/v$ line for data beyond the cut-off for Bragg reflections, λ_c, is $\sigma=(1.9+0.54E^{-\frac{1}{2}})$.

the two elements, there will be a large increase in σ_{inc}/σ_b for the mixture relative to either element alone. If there is some degree of order such that the atoms of the two elements tend to take different regular lattice positions, the value of σ_{inc} is reduced, and this offers a method for the study of such order-disorder effects in favorable cases. In the experiments here described, a state of disorder was assured by rapid quenching of the sample from a high temperature.

Let us consider typical transmission data for a microcrystalline sample of an element as obtained with the neutron velocity spectrometer, and plotted as cross section *vs.* neutron wave-length (see, for example, the curve for nickel given in Fig. 1). This curve may be analyzed as follows: The intercept on the zero wave-length axis is σ_f as given by Eq. (5). The curve between the zero wave-length axis and the first crystal diffraction peak is a straight line whose slope is determined by the capture cross section of the sample. The diffraction effects end at the cut-off wave-length, λ_c, for the crystal, since the portion of the curve beyond λ_c is a region in which there is total destructive interference of the various coherent scattering amplitudes. The curve beyond λ_c is again a straight line parallel to the initial part of the curve, and the intercept obtained by extrapolation of this line to zero wave-length gives σ_{inc}. σ_b is obtained by multiplying σ_f by $(A+1)^2/A^2$, and the effective coherent scattering cross section, σ_{coh}', is obtained from $\sigma_{coh}'=(\sigma_b-\sigma_{inc})$. The latter is smaller than σ_{coh} given by Eq. (7) because of thermal motion of the atoms in the crystal.

TABLE I. Some important observed and calculated cross sections, and the cut-off wave-lengths, λ_c. Cross sections are in barns ($=10^{-14}$ cm²).

	σ_f	σ_b	σ_{coh}'	σ_{coh}, calculated for Same phase	Opposite phase	λ_c in A
Copper	8.2	8.5	6.6			4.16
Manganese	1.80	1.87	~1.9			12.6
Nickel	17.4	18.0	13.9			4.06
Cu–Ni alloy	12.4	12.8	10.1	9.9	0.35	4.11
Mn–Ni alloy	13.6	14.1	5.5	9.5	5.4	6.4

To determine the sign of the scattering amplitude of an element, a substitutional solid solution is formed of the element and another reference element for which the sign of the scattering amplitude is known or assumed. The coherent scattering cross sections for the elements are known from measurements made on the elements

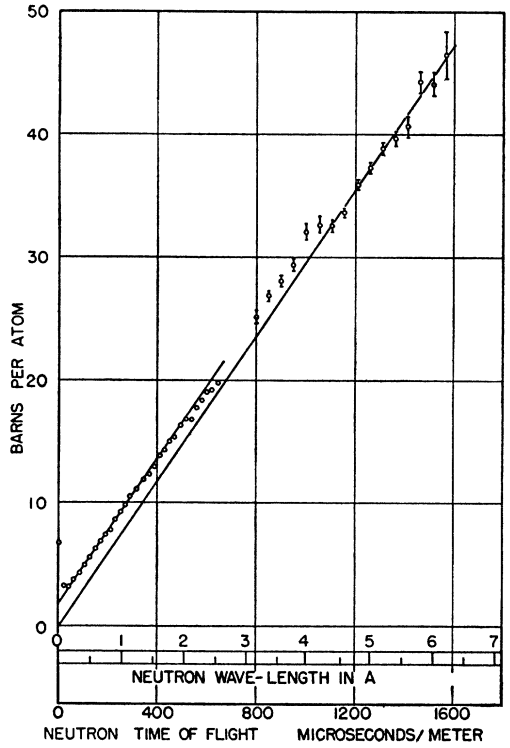

FIG. 3. The slow neutron cross section of manganese as measured with a 16.92-g/cm² sample. The $1/v$ line for the short wave-length is $\sigma=(1.80+2.14E^{-\frac{1}{2}})$.

separately. From a modification of Eq. (7),

$$\sigma_{coh}=4\pi[p_1 f_{1-eff}+p_2 f_{2-eff}]^2. \quad (10)$$

σ_{coh} of the solid solution is calculated assuming f_{1-eff} and f_{2-eff} of the constituent elements to have the same sign, and again assuming opposite signs. A comparison of the measured σ_{coh}' with the two calculated values of σ_{coh} shows whether the phase of scattering of the two elements is the same or opposite.

To be more rigorous, the cross-section data beyond λ_c should be obtained with the sample at a temperature well below the Debye temperature, so as to obtain σ_{coh} rather than σ_{coh}'. However, the difference between these values is small (<0.5 barn) compared with the

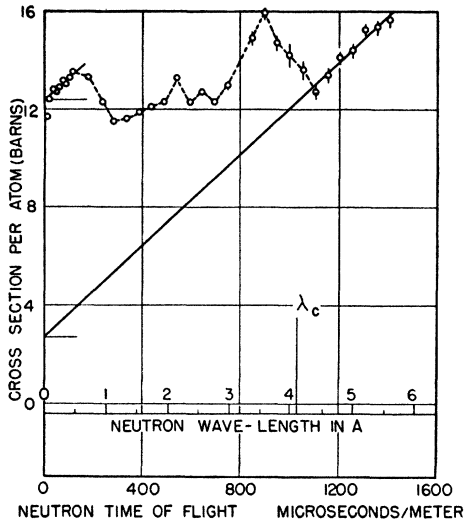

FIG. 4. The slow neutron cross section of disordered copper-nickel alloy composed of 48.10 percent nickel and 51.69 percent copper by weight, and having an area density of 7.48 g/cm². The intercepts of the $1/v$ lines give $\sigma_f = 12.4$ barns and $\sigma_{inc} = 2.7$ barns.

large difference between the values of σ_{coh} calculated for the same and opposite phases of scattering. By the proper choice of the reference element, and of the relative proportions of the constituents of the solid solution, the difference in σ_{coh} for different phases of scattering can be maximized. The scientific literature[9] provides information on a large number of substitutional solid solutions, from which a suitable reference element may be chosen.

EXPERIMENTAL PROCEDURE

To illustrate this method, transmission measurements were made on samples of pure nickel, copper, manganese, and on copper-nickel and manganese-nickel alloys. The nickel sample was composed of seven micron particles of pure nickel obtained by the decomposition of nickel carbonyl. The copper sample consisted of c.p. copper pellets between 2 and 5 mm diameter. The manganese sample consisted of 99.9 percent electrolytic manganese, which was crushed and screened to give a uniform batch of 0.5- to 1-mm particles, and was outgassed at 400°C for 4 hr. in a high vacuum system to remove hydrogen occluded during the electrolytic process. The samples were packed in flat, cylindrical aluminum containers, 3 in. in internal diameter, and $\frac{3}{8}$ to $1\frac{1}{2}$ in. thick, depending on the desired neutron transmission. After careful outgassing, the containers were filled with helium at atmospheric pressure and sealed.[10]

The copper-nickel alloy was in the form of a plate with an area density of 7.48 g/cm², and analyzed 48.10 percent nickel and 51.69 percent copper by weight. It was heated to 800°C and quenched to room temperature with powdered dry ice in less than 1 min. to insure that it was in a completely disordered state. The manganese-nickel alloy consisted of a plate of area density 3.85 g/cm² and analyzed 27.45 atom percent manganese and 72.55 atom percent nickel. The alloy was heated in an inert atmosphere of argon to 800°C and quenched in dry ice to put it in a disordered state.

The operation of the neutron velocity spectrometer has been described in detail by Rainwater and Havens.[11–13] In order to extend the transmission data in the very low energy region (>5A), the source-to-detector distance was reduced from 6 to 5 meters, and a "cold source" consisting of a liquid-nitrogen-cooled paraffin slab was used. For low energy measurements (>3A), the neutron beam was filtered through 13 g/cm² of Be and 11 g/cm² of BeO so as to reduce the stray neutron background.[10] "Standard filters"[14] were used for the high energy measurements to minimize changes in the counting rate for the "sample in" and "sample out" positions.

RESULTS

The cross-section data for nickel, copper, and manganese are shown in Figs. 1, 2, and 3, respectively. These results are considered more accurate and have been extended to longer neutron wave-lengths than the previously published curves for these elements.[1,13] The

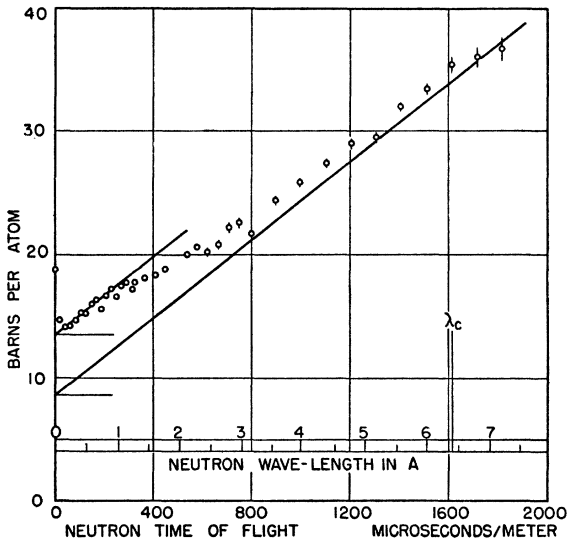

FIG. 5. The slow neutron cross section of disordered manganese-nickel alloy composed of 27.45 atom percent manganese and 72.55 atom percent nickel, and having an area density of 3.85 g/cm². The intercepts of the $1/v$ lines give $\sigma_f = 13.6$ barns and $\sigma_{inc} = 8.6$ barns.

[9] See, for example, *International Critical Tables* (McGraw-Hill Book Company, Inc., New York, 1927), Vol. II, pp. 400–455.
[10] I. W. Ruderman, Phys. Rev. **76**, 1572 (1949).
[11] J. Rainwater and W. W. Havens, Jr., Phys. Rev. **70**, 136 (1946).
[12] W. W. Havens, Jr., and J. Rainwater, Phys. Rev. **70**, 154 (1946).
[13] Rainwater, Havens, Wu, and Dunning, Phys. Rev. **71**, 65 (1947).
[14] E. Melkonian, Phys. Rev. **76**, 1750 (1949).

best $1/v$ line for nickel in the high energy region (0–0.8A) is given by the equation

$$\sigma=(17.4\pm0.2)+(0.79\pm0.1)E^{-\frac{1}{2}},$$

where σ is the total cross section in barns ($=10^{-24}$ cm^2) and E is the neutron energy in electron volts. Beyond λ_c, the best $1/v$ line is given by

$$\sigma=(4.1\pm1.2)+(0.79\pm0.1)E^{-\frac{1}{2}}.$$

After correcting the free cross section for binding, the best value of σ_{coh}' is 13.9 barns.

Similarly, the best high energy $1/v$ lines for copper and manganese are given by

$$\sigma=(8.2\pm0.2)+(0.54\pm0.05)E^{-\frac{1}{2}}$$

and

$$\sigma=(1.80\pm0.05)+(2.14\pm0.02)E^{-\frac{1}{2}},$$

respectively. Beyond λ_c the cross section for copper is given by

$$\sigma=(1.9\pm0.6)+(0.54\pm0.05)E^{-\frac{1}{2}}.$$

The manganese curve could not be observed out to cut-off, because the neutron intensity at this long wave-length (12.6A) is at present too weak to be useful. However, since the best straight line with the correct capture slope through the points between 4.5 and 6A passes through zero, the incoherent scattering cross section is probably very small, and was indeed taken to be zero for this work. In Table I the important cross sections and values of λ_c for each sample are tabulated.

Using the data from Figs. 1 and 2, f_{eff} was calculated for nickel and for copper. Substituting these f_{eff} values in Eq. (10) together with the appropriate p's shows that σ_{coh} for the copper-nickel alloy should be 9.9 barns if copper and nickel scatter with the same phase, and 0.35 barn if the elements scatter with opposite phases. The measured value of σ_{coh}' for the copper-nickel alloy (Fig. 4) is 10.1 barns. Copper and nickel therefore scatter neutrons with the same phase, in agreement with Fermi and Marshall's results on the basis of the total reflection of neutrons from mirrors[3]. Their measurements also show that the signs of the scattering amplitudes of copper and nickel are positive.

From the manganese-nickel curve (Fig. 5) σ_{coh}' is found to be 5.5 barns. If the two elements scatter with the same phase, then from Eq. (10) σ_{coh} of the alloy should be 9.5 barns, whereas if the elements scatter with opposite phases, then σ_{coh} should be 5.4 barns. It may therefore be concluded that manganese has a negative scattering amplitude, in agreement with Fermi and Marshall's results on the basis of the intensities of Bragg reflections from MnS_2,[3] and Wollan and Shull's measurements of Bragg reflections from MnO.[4] This experiment would have been more sensitive if the alloy contained 72.55 atom percent of manganese instead of 72.55 atom percent of nickel; however, this would have extended λ_c to 10A, in which region measurements are presently impractical to make because of the low neutron intensity.

The greatest uncertainty in the measurements made here is introduced by the long extrapolation from the points beyond λ_c to zero wave-length, using the capture slope. This means that the latter must be determined with high accuracy, and that enough points beyond λ_c must be obtained to enable an accurate extrapolation to be made. Fortunately the capture slope can be measured with good statistical accuracy at short wave-lengths where the neutron intensity is high, and where crystal diffraction effects are not present. Improvements in the spectrometer are now under way which should make it possible to obtain better statistical accuracy at long neutron wave-lengths.

ACKNOWLEDGMENTS

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