2.9×10^8 ev and 2×10^9 ev is not much different from that shown in Fig. 3 even at much higher altitudes.

To investigate the influence of lateral showers on the results obtained in the present experiments, in the flight on December 17, 1939 the middle counter tube No. 3 was moved out of the line of the remaining counters. Registering for half an hour the lateral showers, the counting rate at an altitude above 5.2 km was found to be 3 percent of that with all the counters in line. This indicates that in our arrangement the lateral showers do not give any appreciable contribution to the observed fourfold coincidences and, therefore, our results are not influenced by them.

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Neutron Studies of Order in Fe-Ni Alloys*

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Neutron transmission measurements are used to study order in Fe–Ni alloys. The difference in neutron transmission between fully annealed and quenched alloys when plotted against the nickel content displays a broad peak around Ni_3Fe and falls to vanishingly small values near 35 atomic percent Ni and pure Ni. The higher the degree of order the greater the neutron transmission. The substitution of 2.3 atomic percent Mo or 4.1 atomic percent Cr for Fe in the annealed 78 atomic percent Fe–Ni alloy caused a decrease in the neutron transmission, relative to the annealed 78 atomic percent Fe–Ni alloy, of 15.6 and 21.2 percent, respectively. The cold working of an annealed binary 75 atomic percent Ni alloy, a treatment known to produce disorder, gave rise to a decrease of 20.6 percent in neutron transmission. These results demonstrate that neutron techniques serve as a useful tool to study order in Fe–Ni alloys, and suggest that they can be extended to study other solid state phenomena.

THE previous work of Whitaker and Beyer[1,2] has shown that slow neutron interaction may be modified by the physical state of the material which the neutrons traverse. The dependence of neutron interaction on physical parameters of the system affords a possibility of investigating solid state phenomena. If only one parameter in a system be varied, changes in the neutron interaction may be correlated with the parameter in question. In the research reported here this approach was employed to investigate order in Fe–Ni alloys.

The measurements were made with neutrons diffusing from paraffin at room temperature. These neutrons have an energy distribution which corresponds to a de Broglie wave-length spectrum with the most probable wave-length in the region of 1.7 angstroms. The spacings of atoms in a crystal are of about this magnitude, so that interference phenomena should occur when these neutrons interact with matter in the solid state.

Although x-rays are very suitable for studying many aspects of the solid state there exist many problems where x-ray studies yield inconclusive results. This is the situation in the case of super-structures in alloys with the relative intensity of the super-structure lines depending on the difference in scattering powers of the constituent atoms.

In general, x-ray scattering is proportional to the number of electrons in the atoms (unless near

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

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

an absorption edge) and the addition of one unit of charge to the nucleus and one electron to an outer shell produces only a slight change in the scattering power. On the other hand the addition of an extra particle to the nucleus may completely change the response of the system to a slow neutron and neighboring elements in the periodic table may not only differ greatly in their scattering power for neutrons but important phase shift changes in the scattered wave may occur.

The presence of an ordered phase in alloys[3, 4] can be established (or at least inferred) from a study of several physical properties such as electrical conductivity, the effect of plastic deformation on electrical conductivity, specific heats, thermal expansitivity, magnetic properties, etc. At the beginning of this work there was no direct unequivocal evidence, such as x-ray super-structure lines, for the presence of an ordered structure in Fe–Ni alloys. It was also of much interest to determine whether or not the presence of "short range order," i.e., ordered regions approaching the size of two atoms along an edge could be detected from neutron scattering. The most reliable evidence[5] indicates that x-ray super-structure lines are not detectable in Cu_3Au when the ordered domains are of a size less than 16–18 atoms along an edge. There is the further possibility that the presence of out-of-step domains may complicate the situation and prevent the appearance of super-structure lines even though the individual domains were sufficiently large.

TABLE I. *Effect on relative transmissions of the addition of a third component to the* Fe–Ni *alloys.*

ALLOY	RELATIVE TRANSMISSION
78 Fe–Ni annealed	0.500
4.1 Chromium annealed*	0.422
2.3 Molybdenum annealed*	0.394
78 Fe–Ni cold worked	0.397
4.1 Chromium cold worked	0.402
2.3 Molybdenum cold worked	0.369

* The annealing conditions were chosen to develop high initial permeability as discussed above.

PREPARATION OF ALLOYS

A series of iron-nickel alloys including pure iron and nickel were prepared from carbonyl iron and high purity electrolytic nickel in a vacuum induction furnace. The alloys were then subjected to a homogenizing anneal at 1000 degrees C. Pairs of samples were prepared from the cast and homogenized ingots which were 4 by 4 by 0.31 cm in size. All samples were then annealed in a vacuum induction furnace at about 900 degrees C in order to establish comparable grain size in both specimens. One sample of each composition was then annealed in a vacuum induction furnace containing low vapor pressure oil in the bottom of the vacuum chamber. The specimens were suspended on a fine tungsten wire which was fused at the end of the annealing period permitting the samples to drop into the oil bath. This drastic quench should suppress all ordering processes possessing a relaxation time greater than a fraction of a second. The remaining specimens of each of the pairs were then placed in evacuated glass tubes and subjected to prolonged annealings in order to permit the establishment of a large degree of order. They were held at a temperature of 490 degrees C for a period of 700 hours and then slowly cooled to 360 degrees C over a period of 340 hours. This treatment should be suitable for establishing a very high degree of order in the 75 atomic percent Ni alloy and should also produce a high degree of order in samples of other compositions from 85 to 45 atomic percent Ni. Unfortunately very little information is available concerning the optimum ordering temperatures in alloys other than Ni_3Fe, consequently it is extremely difficult to obtain comparable degrees of order as a function of composition. From studies of electrical conductivity of Fe–Ni alloys to be reported later we know that both quenched and annealed alloys are ordered, the difference being in the degree of order.

The Fe–Ni–Cr and Fe–Ni–Mo samples were prepared in a high frequency induction furnace and cold rolled from a plate $\frac{3}{4}$ in. in thickness down to a $\frac{1}{4}$ in. plate. Magnetic measurements show this treatment to give initial permeabilities of 492 and 178, respectively. A suitable heat treatment followed by slow cooling provoked an

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[3] F. C. Nix and W. Shockley, Rev. Mod. Phys. **10**, 1 (1938).

[4] F. C. Nix, J. App. Phys. **8**, 783 (1937).

[5] C. Sykes and F. W. Jones, Proc. Roy. Soc. London **157**, 213 (1936).

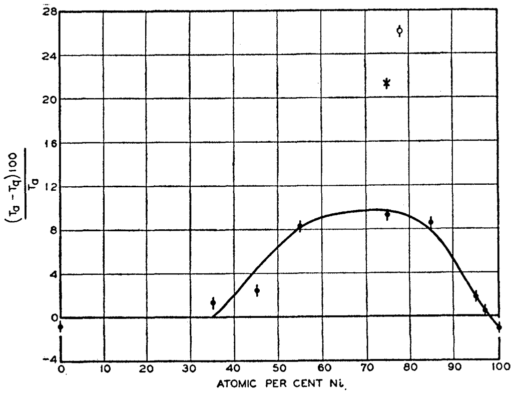

FIG. 1. Difference in neutron transmission *versus* composition (·). T_a, transmission of annealed plate; T_q, transmission of quenched plate. Cold worked plate relative to an annealed plate (*x*). 2.3 atomic percent Mo, Fe–Ni–Mo cold worked plate, relative to annealed 78 atomic percent Ni, Fe–Ni alloy (*o*).

increase in the initial permeabilities to 5400 and 15,100, respectively.

EXPERIMENTAL TECHNIQUE

The general arrangement for the neutron measurements is the same as used previously in the Columbia laboratories.[2]

Neutrons from a 500 to 750-mC Rn–Be source were slowed in a paraffin "howitzer" at room temperature. The emerging neutron beam was highly collimated by a cadmium lined tubular shield filled with boron carbide.

The detector consisted of a BF_3 ionization chamber connected to a linear amplifier and scaling system recorder. Samples to be tested were placed at the midpoint of the neutron beam. Under these geometrical conditions neutrons scattered by the sample through angles greater than 4 degrees did not reach the detector.

Method of observations

In these experiments it was more important to measure accurately the relative difference in neutron transmission between two samples, rather than the absolute value of the neutron transmission, since the samples had the same chemical composition and geometrical shape and differed only in that one of each pair had been quenched and the other annealed as described above. Hence in general, only cyclic measurements of the relative neutron transmission were made, although in some cases absolute transmissions were determined, as shown in Table II. The thicknesses of the samples were the same and in general gave an average neutron transmission of approximately 50 percent.

Precision.—30,000 to 150,000 neutrons were generally counted for each sample so that precision of the determinations of the difference in transmission was less than 1 percent.

Results

The transmission differences between annealed and quenched alloys were studied as a function of the nickel content. Pure iron and nickel were included as a check on the experimental technique of the preparation of the samples. Representing by T_q and T_a the transmission of the quenched and annealed plates, respectively, the values of $(T_a - T_q)100/T_a$ for various concentrations of Ni are:

At. % Ni	100	97	95	85	75	55	45	35	0
$100(T_a - T_q)/T_a$	−0.9	0.4	1.8	8.5	9.3	8.2	2.5	1.3	−0.6

The effects of cold working by compression and of quenching on the neutron transmissions were also ascertained for comparison. The relative transmissions of the annealed, quenched, and cold worked 75-atomic percent Ni samples are:

TREATMENT	ANNEALED	QUENCHED	COLD WORKED
Transmission	0.500	0.454	0.397

The effect of adding a third component to the Fe–Ni alloys was studied with plates containing 4.1 atomic percent Cr and 2.3 atomic percent Mo substituted for some of the Fe, in the 78 atomic percent Ni alloy. Mo and Cr have smaller slow neutron cross sections than Fe, the values being 7×10^{-24} cm^2 and 6×10^{-24} cm^2 as contrasted

TABLE II. *Observed neutron cross sections for a series of* Fe–Ni *alloys.*

	CROSS SECTION $\times10^{24}$ CM^{-2}		
ATOMIC PERCENT Ni OF Fe-Ni ALLOYS	ADDITIVE (EXPECTED)*	OBSERVED	PERCENT CHANGE
78 (annealed)	18.2	12.5	31.3
78 (quenched)	18.2	15.5	14.8
78 (cold worked) (containing 2.3 at. % Mo)	18.1	18.0	0
45	15.6	15.5	0

* Additive values computed from neutron cross sections of polycrystalline Fe, Ni and Mo.

with 12×10^{-24} cm^2 for Fe. The relative transmissions are given in Table I.

The results are graphically presented in Fig. 1.

Table II shows the observed neutron cross sections for a series of Fe–Ni alloys together with the cross sections assuming additivity.

Discussion of Results

The differences in neutron transmission between the quenched and annealed alloys can be measured at compositions where super-structure x-ray lines cannot be detected even by refined x-ray technique. Super-structure lines have been detected only in 70[6] and 75[6] atomic percent Ni alloys,[7,8] whereas reliable differences in neutron transmission can be measured in the entire composition region from 45 to 90 atomic percent Ni. It is possible that neutrons reveal the presence of considerably smaller ordered domains than are disclosed by x-rays; this could account for the different results obtained by the two methods.

It has also been shown[6] that cold working of both annealed and quenched Fe–Ni alloys around the Ni_3Fe region provokes an increase in the electrical resistivity. Cold working of an ordered Cu_3Au alloy gives rise to a similar increase in electrical resistivity, and here the accompanying disappearance of the x-ray super-structure lines with cold working in the latter alloy shows that the increase in the electrical resistivity is correlated with the destruction of order. The results of neutron transmission studies indicate that a similar correlation occurs in Fe–Ni alloys. Thus the neutron results lead to the conclusion that both quenched and annealed Fe–Ni alloys in the region around Ni_3Fe are ordered, the difference being simply in the degree of order.

In the case of the ternary alloys containing 4.1 atomic percent Cr and 2.3 atomic percent Mo the neutron transmissions of both cold worked and annealed alloys are appreciably smaller than that of the binary Fe–Ni alloy of the same Ni content, indicating a lesser degree of order in the ternary alloys. It is also of interest to note that a heat treatment suitable to increase the initial permeability from 178 to 15,100 in the Mo–Fe–Ni alloy changes the neutron transmission difference, relative to the binary Fe–Ni alloy, only from 26.2 percent to 21.2 percent. This heat treatment was such as to cause some increase in the degree of order; however, the small change observed in neutron transmission could not be accounted for by the large change in initial permeability.

It should be pointed out that magnetic polarization effects do not play any appreciable part in the neutron transmission changes observed in these researches. The Fe–Ni–Mo alloy, which has the highest initial permeability, should be most transparent from magnetic polarization considerations, whereas actually this alloy scatters more neutrons than the other alloys. Furthermore; the transmission changes reported in this paper are very much larger than observed with magnetic polarization effects and, in addition, neutron transmission changes due to magnetic polarization have only been observed with very high magnetizing fields.[9]

The results are very consistent, with the assumption that changes in degree of order affect the neutron transmission, in that where large deviations from additivity occur, the thermal treatments and cold working, which are known to produce changes in order, are effective in changing the neutron transmission.

Conclusions

These results clearly show that neutron transmission measurements provide a very sensitive technique for studying changes of order in Fe–Ni alloys. Such measurements are able to give quantitative comparisons of the effect of various physical processes, such as cold working and thermal treatments, in changing the state of order.

In the particular case of Fe–Ni alloys studied in these experiments, the sensitivity of the neutron transmission technique may be due in part to large phase-shift differences in the waves scattered by the Fe and Ni nuclei. If this is true, then the method will not be equally sensitive for all alloys, but it should be applicable to many.

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[8] P. Leech and C. Sykes, Phil. Mag. **27**, 742 (1939).

[9] J. R. Dunning, P. N. Powers and H. G. Beyer, Phys. Rev. **51**, 51, 371 (1937).