

with a relatively low average field. Comparison with the magnetization data of Wakelin and Yates<sup>8</sup> shows that the composition difference, found from Fig. 4, can account for this increase.

The observed hysteresis is certainly not self-evident. First of all it may be noticed that the hysteresis zone is not symmetrical around the two-phase region. So the disordering starts immediately when the material is heated to a temperature inside or above the two-phase region. On the other hand the ordering reaction is suppressed and starts only more than 10 K below the two-phase region. It should be noted that in contrast to disordering no nucleation centers are available for the ordering process. The suppression of the nucleation is probably of magnetic origin, which is also suggested by the maximum in the transition temperature at 27 at. % Fe. Further evidence may be found in the absence of hysteresis in Cu<sub>3</sub>Au,<sup>11</sup> a material which has properties similar to Ni<sub>3</sub>Fe, but which is not ferromagnetic.

In conclusion, we have shown that Mössbauer spectroscopy provides us with a valuable tool for the study of order-disorder phenomena in Ni<sub>3</sub>Fe. Both the microscopic nature of the measuring technique and the possibility to follow the reactions "at temperature" facilitate the interpretation of the results. Three transition mechanisms were observed in the transition region: nucleation and growth both with and without compositional segregation, and a further homogeneous growth of order inside ordered segregates. These

results, together with the hysteresis zone, are fully consistent with a classical first-order transition. Our research will be extended to other compositions, especially to the 27-at. % Fe composition where, on the basis of the phase diagram, a direct transition is expected. In order to study the spatial character of the observed phenomena, a concurrent electron-microscopical investigation is in progress.

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## Magnetic Excitations in the Amorphous Ferromagnet Co<sub>4</sub>P†

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Magnetic excitations in the amorphous ferromagnet Co<sub>4</sub>P have been studied by use of neutron-inelastic-scattering techniques. Measurements around  $Q = 0$  show distinct spin-wave excitations that obey a quadratic dispersion law  $E = DQ^2$  where  $D = 185 \text{ meV } \text{Å}^2$ . Additional measurements around the first peak in the static structure factor  $S(Q)$  show magnetic excitations that appear analogous to the roton excitations in <sup>4</sup>He. These excitations are discussed in terms of sum rules for the magnetic scattering.

Neutron-inelastic-scattering techniques have been used to measure the magnetic excitations in the amorphous ferromagnet Co<sub>4</sub>P. A distinct

spin-wave excitation with a quadratic dispersion law was measured around momentum transfer  $Q = 0$  by use of a triple-axis spectrometer. Fur-

their measurements at  $Q$  values near the first peak in the static structure factor  $S(Q)$  showed an additional part of the excitation spectrum that has a minimum at the peak of  $S(Q)$  and rises on either side of this minimum in an almost quadratic manner. These excitations appear to be similar to the roton excitations in  $^4\text{He}$  and will be discussed in terms of sum rules on the magnetic scattering. From the sum rules we will be able to derive a relation for the excitation spectrum near the roton minimum similar to that obtained by Feynman for the case of  $^4\text{He}$ .<sup>1</sup>

The sample used in the present measurements was made at Harvard University and has a ferromagnetic Curie temperature of about  $620^\circ\text{C}$  and a moment of about 1.0 Bohr magneton per cobalt atom at room temperature, which is the temperature at which the measurements were taken. The material has the usual liquidlike static structure factor  $S(Q)$  that one would expect for an amorphous material with the exception that the first peak in  $S(Q)$  occurring at  $3.13 \text{ \AA}^{-1}$  is somewhat higher and sharper than is found in most structurally disordered systems. The radial distribution function for the material obtained from  $S(Q)$  is similar to that obtained for a dense random packing of hard spheres and the average metal-metal near-neighbor separation is only about 2% larger than the distances of closest approach in the hcp crystalline form of cobalt.<sup>2</sup>

It is not surprising that there should be long-wavelength spin waves in such a material since the long-wavelength modes would not be too dependent on the local order. Figure 1 shows triple-axis-spectrometer scans near  $Q=0$  that show distinct spin-wave excitations. The excitations appear to broaden at higher momentum transfers, but in reality the broadening is due to the fact that the resolution ellipse of the spectrometer is taken through a dispersion curve whose slope is becoming greater at higher energies and the spin-wave width is consistent with that expected from spectrometer resolutions alone. The measurements were made with an incident energy of 4.7 meV and were difficult since the scattering angles involved were less than a degree in most cases. This meant that good resolution in  $Q$  and  $E$  was necessary. Fortunately, it was possible to just resolve the spin wave before the scattering angle became too small to avoid scattering from the direct beam; the rapid increase in neutron intensity at the end of the scans is due to the collapse of the scattering triangle. Only very limited mea-

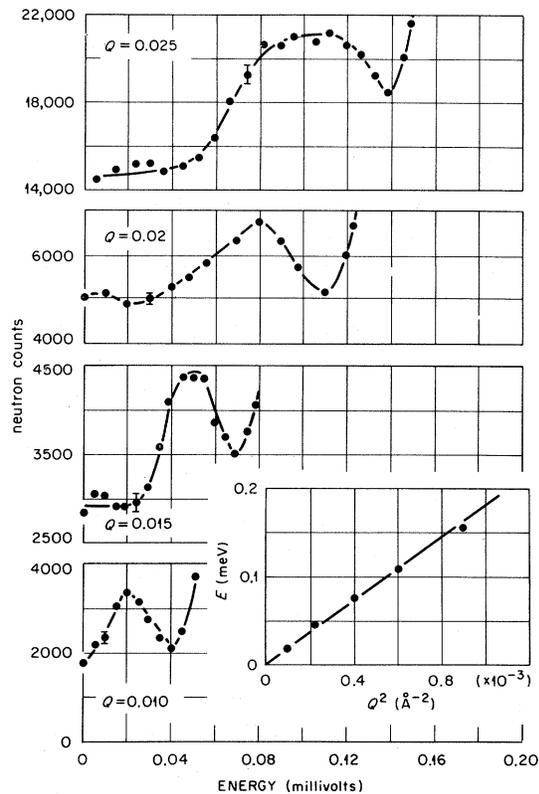


FIG. 1. Magnetic excitations near  $Q=0$  measured with a three-axis spectrometer.

surements, up to  $Q=0.03 \text{ \AA}^{-1}$ , were possible before the dispersion curve became so steep that the  $\Delta E$  needed for the measurement became too high to be measured with the  $Q$  needed to stay on the dispersion curve. If one could obtain an amorphous ferromagnet with a less steep dispersion curve, measurements could be made at higher values of  $Q$ . Nevertheless sufficient data were obtained to plot a dispersion curve and Fig. 1 shows a plot of  $Q^2$  for the spin waves versus their energy (corrected for resolution effects). The resulting straight line shows that the spin-wave dispersion is quadratic and can be fitted by the relationship  $E=DQ^2$  where  $D$  is  $185 \text{ meV \AA}^2$ . This value of  $D$  is inconsistent with the value of about  $120 \text{ meV \AA}^2$  obtained from low-temperature-magnetization measurements by Cochrane and Cargill.<sup>2,3</sup> The cause of this inconsistency is unknown; however, a similar inconsistency was obtained between the magnetization and neutron-scattering measurements for the amorphous ferromagnet  $\text{Fe}_{0.75}\text{P}_{0.15}\text{C}_{0.1}$ .<sup>4</sup> One would expect the neutron-scattering results to be quite reliable since the spin-wave excitations are observed di-

rectly.

At larger values of  $Q$ , measurements can be made at larger energy transfers since one can again satisfy energy and momentum conservation. Measurements were made with a time-of-flight spectrometer that employs a polarized beam.<sup>5</sup> The experiment was arranged so that only spin-flip excitations were observable and the autocorrelation technique was used to obtain a good signal-to-noise ratio. The polarized-beam time-of-flight technique is an excellent method of examining amorphous ferromagnets since one avoids all the phonon and elastic scattering and a large number of momentum transfers can be measured simultaneously by use of multiple detectors.

Magnetic excitations were observed at  $Q$  values near the first peak in  $S(Q)$  at  $3.13 \text{ \AA}^{-1}$ . They were at rather high energies and long counting times (about a week) were needed to get good quality measurements. The excitations were quite sharp in energy which is surprising since excitations of these wavelengths should be quite sensitive to the disorder of the system. The measurements are shown in Fig. 2. The scans are time-of-flight scans in which  $Q$  and  $E$  vary simultaneous-

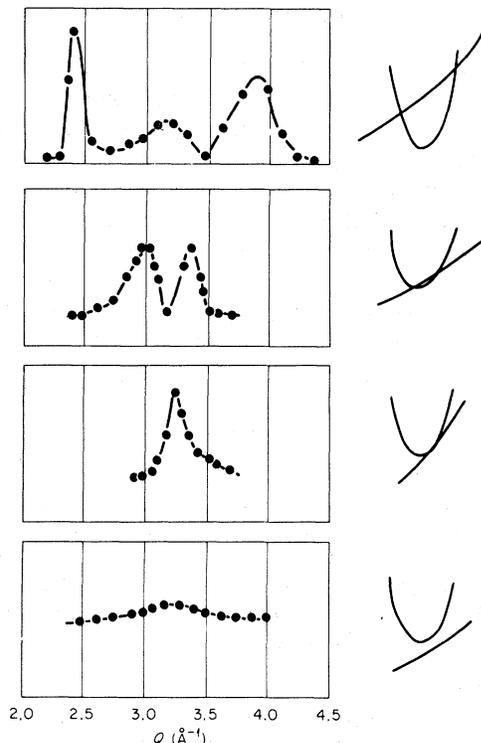


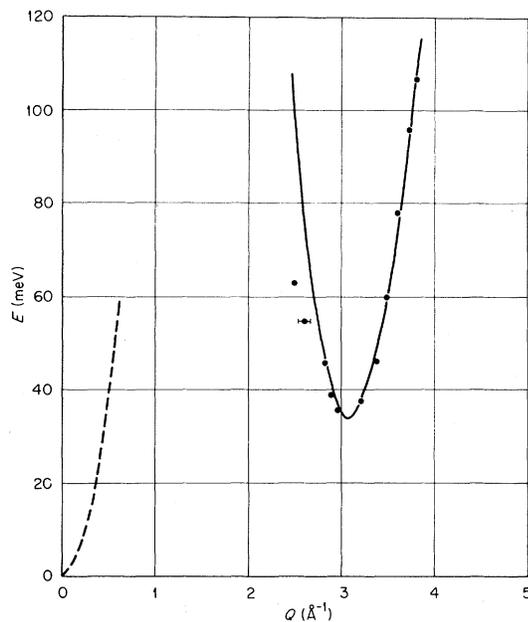
FIG. 2. Magnetic excitations in the roton region near the first peak in  $S(Q)$ .

ly; however, since the energies are rather high, the scans are more nearly constant- $E$  in nature. The top scan is at an average energy of about 80 meV and contains three peaks, the central peak being at  $3.13 \text{ \AA}^{-1}$ , right at the peak of  $S(Q)$ .

As the energy is lowered the three-peak pattern merges into two strong peaks, the central peak probably being hidden by the strong side peaks. Finally the peaks merge into one strong peak at 34 meV and at energies below this one sees nothing but a small bump at  $3.13 \text{ \AA}^{-1}$ . The origin of the small middle peak that remains at  $3.13 \text{ \AA}^{-1}$  independent of energy is unclear. It quite possibly is multiple spin-wave scattering since one would expect this to peak up at the peaks in  $S(Q)$ . The remaining strong peaks lie very nearly on a parabola and the diagrams on the right-hand side of Fig. 2 show the time-of-flight trajectories through the parabola that would fit the observed measurements. The linewidths are again instrumental in nature and no intrinsic broadening of the excitations was observed. An effort was made to measure the intrinsic width of the excitations by making a constant- $Q$  scan at  $3.13 \text{ \AA}^{-1}$  using a triple-axis spectrometer. A peak was observed at 34 meV in agreement with the time-of-flight data. However, the peak was on a sloping background that increased at high energies making it somewhat difficult to extract a width accurately, and, of course, phonon excitations cannot be avoided with the triple-axis spectrometer. Nevertheless, by folding the scan with the spectrometer resolution an upper limit of 5 meV can be put on the spin-wave width at the bottom of the parabola at 34 meV.

Figure 3 shows the spin-wave excitation spectrum if we neglect the small peak at 34 meV that is independent of  $Q$ . Only a small part of the parabola around  $Q=0$  could be measured, but if the data are extrapolated to high energies the dashed line is obtained. The full line shows this same parabola placed with its bottom at  $Q=3.13 \text{ \AA}^{-1}$  and  $E=34 \text{ meV}$ . There is no compelling reason to expect the data to fall on this parabola; however, the data points fall very close to it especially at larger  $Q$ .

The excitation spectrum is quite reminiscent of the phonon spectrum for  $^4\text{He}$ , the phonons of course being linear at small  $Q$ . It is interesting to see how far the analogy between the two systems can be carried, especially around the roton region that occurs at the peak of the static structure factor. Using a series of elegant physical arguments, Feynman<sup>1</sup> deduced that the excitation

FIG. 3. Spin-wave dispersion curve for Co<sub>4</sub>P.

spectrum in the roton region is given by

$$E(Q) = Q^2/2mS(Q), \quad (1)$$

where  $m$  is the <sup>4</sup>He mass. If one postulates a sharp excitation spectrum the Feynman result can be obtained by making use of the  $f$ -sum rule on the scattering law  $S(Q, E)$ ,

$$\int_0^\infty dE S(Q, E)E = Q^2/2m; \quad (2)$$

then with  $S(Q, E) = S(Q) \delta(E - E(q))$  Eq. (1) is obtained immediately.

The quantity of importance in the spin-wave case is the imaginary part of the generalized susceptibility that gives the spin-flip scattering. This quantity is denoted by  $\chi_{-+}''(E, Q)$  and the neutron cross section for spin-flip scattering can be obtained directly if this quantity is known.<sup>6</sup>

The sum rules of importance for  $\chi_{-+}''(Q, E)$  are<sup>7,8</sup>

$$\int_0^\infty dE \chi_{-+}''(Q, E)E = Q^2/4m_e, \quad (3)$$

$$\int_0^\infty dE \chi_{-+}''(Q, E)/E = \chi(Q), \quad (4)$$

where  $m_e$  is the electron mass. Since the observed excitations are sharp it is not unreasonable to postulate a sharp excitation spectrum for a given  $Q$ , and thus making this assumption one obtains immediately from (3) and (4) the dispersion relation

$$E^2(Q) = Q^2/4m_e \chi(Q) \quad (5)$$

which is analogous to Eq. (1).

The above arguments are only qualitative and not quantitative in nature. Equation (1) does not give the roton gap correctly for <sup>4</sup>He and many-body corrections must be applied to the system. Equations (3) and (4) give the sum rules for all of  $\chi_{-+}''(Q, E)$ , including Stoner modes which we have neglected, so that one could not expect (5) to be an exact relationship for the spin-wave excitations. Nevertheless Eq. (5) can certainly give us some important information about the excitation spectrum. We expect  $\chi(Q)$  to be proportional to  $Q^{-2}$  for a ferromagnet as  $Q$  goes to zero so that (5) gives the correct quadratic relationship in this region. Since we expect  $\chi(Q)$  to have peaks in the same place as  $S(Q)$ ,<sup>9</sup> Eq. (5) predicts that the roton minimum is at  $3.13 \text{ \AA}^{-1}$  and predicts a gap, although we cannot determine its value since we lack detailed information about  $\chi(Q)$ . The sum rules are thus quite helpful since we lack detailed theories for excitations in amorphous systems.

The excitation spectrum for phonons in an amorphous system has been arrived at in the quasicrystalline approximation<sup>10</sup> and this can easily be altered to apply to the magnetic system, resulting in the following equation:

$$E(Q) = \sum_{\vec{r}} J(r)g(r)(1 - e^{i\vec{Q}\cdot\vec{r}}), \quad (6)$$

where  $J(r)$  is an exchange function and  $g(r)$  is the pair distribution function. This approximation is far too crude to be realistic, however, and we have found that Eq. (6) cannot be made to fit the observed data regardless of how one chooses  $J(r)$ . We hope then that our results will encourage further theoretical work on excitations in amorphous systems.

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