

The Space Lattice and "Superlattice" of Pyrrhotite

Both paramagnetic and ferromagnetic solid solutions of pyrrhotite ($\text{FeS}_{1.05}$ – $\text{FeS}_{1.12}$) may be found which are outwardly at least structurally isomorphous. The latter has anomalous ferromagnetic and gyromagnetic properties. In solutions from 50.0 to 51.2 mole percent sulfur (corresponding to $\text{Fe}_{19}\text{S}_{20}$) however, a "superlattice," in addition to the basic hexagonal lattice, has been reported.¹ As reported, the symmetry of the "superlattice" is also hexagonal, but the (110) axis of the old elementary cell (of the nickel arsenide type) becomes the a axis of the super cell, while the c axis of the old cell is doubled in length. At about 51.6 mole percent sulfur the "superlattice" is said to disappear and ferromagnetism to appear. The relation of the "superlattice" to ferromagnetism has been further investigated by us through x-ray studies of single crystals, as well as of powder specimens of both ferromagnetic and paramagnetic pyrrhotite.

The evidence for the presence of a "superlattice" in paramagnetic pyrrhotite has been confirmed by means of powder diffraction spectra made with filtered chromium radiation of samples of natural and synthetic pyrrhotite.² Further, identically the same evidence has been obtained from both natural and synthetic ferromagnetic pyrrhotites.² The diffraction lines reported¹ for the "superlattice" are found present in varying number in the diffraction patterns of all the specimens studied.

X-ray diffraction studies have been made on a ferromagnetic single crystal from Minas Geraes, Brazil,³ and on a paramagnetic single crystal from Maggione, Italy.⁴ In addition to Laue photographs, rotation photographs about the a , c , and (110) axes have been prepared with filtered molybdenum radiation. These photographs lead to a hexagonal lattice with the following dimensions:

$$a_0, 3.453 \pm 0.009\text{A}; c_0, 5.670 \pm 0.020\text{A}; \\ d_{110}, 5.976 \pm 0.029\text{A}; c/a, 1.644.$$

The same values of the parameters, within the experimental error, were found for both the ferromagnetic and the paramagnetic crystals.

All the diffractions observed from the single crystals, without exception, were identified as due to reflections from the planes of the basic lattice. In contrast to the powder diffraction data, diffraction spots corresponding to the reflections from the planes of the "superlattice" alone were not observed on the rotation photographs of either the ferromagnetic or the paramagnetic crystals, although from the point of view of intensity it was found possible for them to appear. Hence, this evidence does not support the existence of a "superlattice" in the single crystals of pyrrhotite studied in this investigation.

The Laue photographs of both crystals appear to have the symmetry D_{6h} . Diffractions from the general planes (hkl) appear in all orders. Characteristic halvings are (00 l) and (hhl). Accordingly, the most probable space groups are D_{6h}^4 , C_{6v}^4 , and D_{3h}^4 .

The density of certain powder specimens has been determined by us as 4.55 g/cc. Using this value, the number of molecules of pyrrhotite per elementary cell is approxi-

mately 2. The space groups D_{3d}^4 and C_{3v}^4 , which give the same characteristic halvings as those found, have not been considered, since, when the number of equivalent points is only 2, these groups reduce to two of those given above as far as the atomic positions are concerned; further, these two groups are likely eliminated by virtue of the Laue symmetry observed.

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¹ G. Hägg and I. Sucksdorff, Zeits. f. physik. Chemie **B22**, 444 (1933).
² The samples of natural pyrrhotite were obtained from Kisbanya, Roumania, and Rossland, British Columbia. The synthetic pyrrhotites were kindly supplied by Dr. H. S. Roberts, Geophysical Laboratory, Washington, D. C.

³ Kindly supplied by Professor H. Leighton, department of geology, University of Pittsburgh.
⁴ Kindly supplied by Dr. F. H. Pough, The American Museum of Natural History, New York.

Further Remarks on the Saturation Property of Nuclear Forces

Certain restrictions are imposed on the relative amounts of the different kinds of exchange forces in the general symmetrical interaction operator by the requirement that the binding energy per particle in heavy nuclear systems falls below a constant upper bound which is independent of the number of particles.¹ In particular this condition must be satisfied when the statistical method is used to calculate the binding energies of heavy nuclear systems containing (1) equal numbers of neutrons and protons with zero total spin, (2) neutrons only with zero total spin, (3) equal numbers of neutrons and protons with all spins parallel, (4) neutrons only with all spins parallel. Applied to the interaction operator¹

$$V = \sum_{i < j} \{ W + MP_{ij} + BQ_{ij} + HP_{ij}Q_{ij} \} J(r_{ij}), \quad (1)$$

with $W + M + B + H = 1$, the requirement yields the set of simultaneous inequalities

$$\begin{aligned} c_1 &\equiv 4W - M + 2B - 2H \leq 0, \\ c_2 &\equiv 2W - M + B - 2H \leq 0, \\ c_3 &\equiv 2W - M + 2B - H \leq 0, \\ c_4 &\equiv W - M + B - H \leq 0, \end{aligned} \quad (2)$$

each obtained from the correspondingly numbered type of nuclear system. The condition on c_4 is not independent, but follows from $c_3 \leq 0$. The conditions on c_1 and c_2 were first derived in reference 1. Kemmer² has recently given the third inequality. If, for reasons which have been discussed elsewhere,³⁻⁵ we take $c_1 = 0$, the remaining inequalities reduce to

$$M + 2H \geq 0, \quad M - 2B \geq 0. \quad (3)$$

An independent restriction,⁶ $M - 2B \geq H$, is required by the instability of odd-odd nuclei heavier than N^{14} .

From $c_1 = 0$, Kemmer's inequality, and the relation $W + M - B - H \sim 0.5$ fixed by the scattering cross section

of slow neutrons in hydrogen there follows

$$\begin{aligned} W-M+B-H &= -(W+M+B+H)/3 + 2c_3/3 \leq -1/3, \\ W-M-B+H &= -3(W+M-B-H) + 3(M-2B) \\ &\geq -3/2. \end{aligned} \quad (4)$$

These inequalities put one-sided limits on the triplet and singlet p interactions involved in the elastic scattering of fast neutrons and protons in hydrogen. Furthermore

$$\begin{aligned} 9(W-M+B-H) + (W-M-B+H) \\ = -6(W+M) \sim -9/2, \end{aligned} \quad (5)$$

a useful relation establishing a connection between the p scattering of protons in hydrogen (associated with the triplet interaction) and the p scattering of neutrons in hydrogen (involving both singlet and triplet interactions). It is worth noting explicitly that the upper bound on the triplet p interaction given in (4) is a consequence of $c_3 \leq 0$ alone while Eq. (5) follows from $c_1 = 0$ alone.

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September 2, 1937.

¹ G. Breit and E. Feenberg, Phys. Rev. **50**, 850 (1936).

² N. Kemmer, Nature **140**, 192 (1937).

³ D. R. Inglis, Phys. Rev. **51**, 531 (1937).

⁴ E. Feenberg, Phys. Rev. **51**, 777 (1937).

⁵ H. Volz, Zeits. f. Physik **105**, 537 (1937).

⁶ E. Wigner, Phys. Rev. **51**, 947 (1937). Using the experimental value for the ratio of the triplet and singlet s interactions one finds

$$\begin{aligned} W-M+B-H &= -2(M-2B-H) + (W+M-3B-3H) \\ &\sim -2(M-2B-H). \end{aligned}$$

The β -Ray Spectrum of Mn⁵⁶

We have measured the β -ray spectrum of Mn⁵⁶ excited by bombarding NaMnO₄ solution with neutrons from a Ra-Be source. Using a field of 850 gauss, we obtain a single group with a K-U end-point at 6.5 mc^2 , in agreement with Gaertner, Turin and Crane.¹ Using a field of 425 gauss, we obtain curves similar to those of Brown and Mitchell,² with end-points at 3.4 and 6.5 mc^2 , respectively. With a field of 637 gauss, the spectrum again shows the two groups with the same end-points as found with 425 gauss, but with the relative population of the lower energy group greatly diminished. More than 1300 tracks were measured for each field.

This work then confirms the existence of the low energy group reported by Brown and Mitchell. We attribute the discrepancy between their results and those of Gaertner, Turin and Crane to the suppression of some of the tracks of the low energy group by the stronger magnetic fields, but our work does not permit us to draw conclusions as to the ultimate origin of the low energy group.

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¹ Gaertner, Turin and Crane, Phys. Rev. **49**, 793 (1936).

² Brown and Mitchell, Phys. Rev. **50**, 593 (1936).

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