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AN X-RAY SEARCH FOR THE ORIGIN OF FERROMAGNETISM

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ABSTRACT

A sensitive null method, employing two crystals and two ionization chambers, was used to detect any change in intensity of an x-ray beam reflected from a magnetic crystal, when this magnetic crystal was magnetized. This method was capable of detecting from 1.0 to 0.1 percent change in intensity, according to the intensity of the reflected beam. Crystals of magnetite and silicon steel were used. No change of intensity was observed, although the theoretical change was several times that which could have been detected. The change in intensity of the x-ray beam was computed on the basis of the alteration of the structure factor due to orientation of electronic orbits by the magnetic field. It is concluded that electrons revolving in orbits within the atom cannot account for ferromagnetism. The models of Honda and Ehrenfest are briefly discussed. On the basis of present atomic models and present experimental evidence it appears that the ultimate magnet should be identified with the spinning electron.

SEVERAL experiments have been reported[1] in which x-rays have been used to study the nature of the ultimate magnetic particle. From these it has been concluded that the most minute crystal aggregations in solid iron are not oriented by a magnetic field, that the ultimate magnet cannot be a group of atoms such as the chemical molecule, and finally, that the atom as a unit cannot account for ferromagnetism. On the basis of present atomic models three possible ultimate magnetic particles remain: the atomic nucleus, the spinning electron, or electrons revolving in inner orbits. The following experiment was performed primarily to ascertain whether electrons revolving in the inner orbits may be responsible for ferromagnetism.

THE EXPERIMENT

The apparatus used is indicated in Fig. 1. It is essentially a refinement of that employed by A. H. Compton and Rognley. The x-ray tube had a water-cooled molybdenum target, and could be operated at 35 milliamperes

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[1] M. de Broglie, Le Radium **10**, 186 (1913). K. T. Compton and E. A. Trousdale, Phys. Rev. **5**, 315 (1915). A. H. Compton and Oswald Rognley, Phys. Rev. **16**, 464 (1920). T. D. Yensen, Phys. Rev. **31**, 714 (1928).

and 50 kilovolts. One portion of the x-ray beam fell on C', a crystal of magnetite or silicon steel, which reflected the $K\alpha$ line into ionization chamber I'. Another portion of the beam passed above C' and was incident upon C'', a calcite crystal, from which the $K\alpha$ line was reflected into chamber I''. The electrodes of the ionization chambers were connected to the same pair of electrometer quadrants, while one chamber was maintained at a positive and the other at a negative potential. S' and S'' were adjustable slits. An intense beam of x-rays was allowed to fall upon the crystals, and the slits were adjusted until the electrometer charged up at a very slow rate. With other conditions unaltered the crystal C' was magnetized and the rate of deflection of the electrometer again taken. In order to make a comparison of successive readings, two readings taken with the electromagnet unenergized alternated with two successive readings with the magnet energized. From a set of twenty-five to fifty readings for each order the mean change of rate due to magnetization was determined. A sheet of aluminium, which absorbed four percent of the beam was then placed in front of S' and the resulting change in the rate at which the electrometer was being charged observed. Then: Change of rate due to magnetization/Change in rate due to absorption by aluminium $\times 4 =$ percent change in intensity due to magnetization.

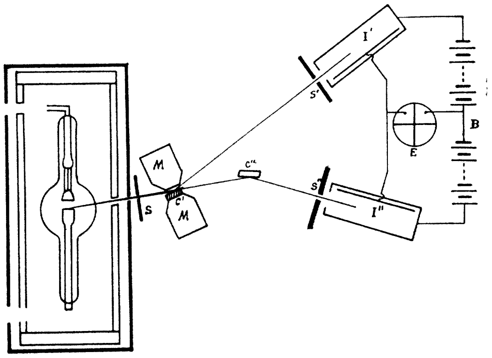

Fig. 1. Diagrammatic sketch of apparatus.

The electromagnet M had a laminated core about which were wound five thousand turns of number 23 copper wire. The air gap was about 2 mm. Demagnetization was secured by successively reversing and gradually decreasing the current. The magnetic induction was measured by means of a commercial flux meter. The calcite crystal was fixed to a non-magnetic mounting which was placed outside the magnetic field and could be rotated independently of the electromagnet and the magnetic crystal. The x-ray tube was mounted inside a heavy wrought iron pipe, the ends of which were closed by iron plates one inch in thickness. The ionization chambers were duplicates, and each was filled with argon.

DISTURBING EFFECTS AND THEIR ELIMINATION

It was necessary to mount the crystal C' with unusual care in order to avoid motion of the crystal relative to the incident beam when the magnet was energized. It was found also that the establishing of the magnetic field sufficient in strength to produce magnetic saturation in the crystal shifted the electrometer needle through several degrees, and changed the current sensitivity of the electrometer. By using a mica needle plated with a suitable combination of gold and platinum this effect was overcome. In testing the sensitivity the deflection rates were compared with and without the magnetic field, using radium emanation tubes as a source of ionization.

Before the tube was enclosed in the iron pipe, the field in the region of the x-ray tube was found to be sufficient to move the focal spot 0.2 or 0.3 mm. This shift produced a change in the intensity of the reflected x-rays of the order of 1 percent, which changed in sign with a reversal of the field. When the tube was placed in the iron pipe, tests showed the shielding to be adequate.

CALCULATION OF CHANGE IN REFLECTION DUE TO ORIENTATION OF AN ELECTRON ORBIT

The following method was used in computing the theoretical change in the intensity of the reflected beam due to K or L electrons being oriented by an external field so that their magnetic moments were parallel to this field. A. H. Compton[2] shows that

$$W_n = KF_n^2, \tag{1}$$

where W_n is the total energy diffracted by a small crystal and F_n is the structure factor of the atom, while the subscript n refers to the order in question, and K is a factor of proportionality. If the primed letters refer to the energy and structure factor respectively when the crystal is magnetized and the unprimed letters to the same quantities when the magnetic crystal is unmagnetized, then

$$\frac{W'}{W} = \frac{(F_n')^2}{(F_n)^2} = \frac{[F_n + (F_n' - F_n)]^2}{(F_n)^2} = 1 + \frac{2\Delta F_n}{F_n} \cdot \tag{2}$$

neglecting higher powers of ΔF_n. To calculate the change in intensity it is thus necessary to know the number of atoms affected by the magnetic field and the structure factors of the atom for the order in question when the crystal is magnetized and unmagnetized respectively.

The structure factor[3] for the nth order of an electron moving at random over the surface of a spherical shell of radius r is

$$F_{en} = \frac{\sin 2\pi nr/D}{2\pi nr/D}, \tag{3}$$

where D is the grating space of the crystal. If magnetizing the crystal perpendicular to the reflecting face causes the orbit of an electron to lie in the plane of the reflecting face, its structure factor becomes unity, since the waves reflected by it are in phase with those from the middle of the atomic

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[2] A. H. Compton, "X-rays and Electrons" p. 125.

[3] Reference 2, p. 148.

layer. If without the magnetic field the orientation of the orbit is random, then

$$\Delta F_{en} = 1 - \frac{\sin 2\pi nr/D}{2\pi nr/D} \cdot \tag{4}$$

If, however, the crystal is magnetized parallel to the reflecting face, and the electron in question moves in a circle whose plane is perpendicular to the reflecting face, the structure factor for this electron becomes a Bessel function of the zero order,

$$F_{en}' = J_0(2\pi nr/D) . \tag{5}$$

ΔF_{en} is then found in the same manner as in the preceding case.

If ΔF_{en} is the change in structure factor for each electron whose orbit is turned by the magnetic field, and if f is the number of electronic orbits per atom thus turned, the average change in structure factor is

$$\Delta F_n = f\Delta F_{en} . \tag{6}$$

From Eq. (2) it follows that the ratio of the intensities when the crystal is magnetized and unmagnetized respectively is

$$(W'/W) = 1 + 2f(\Delta F_{en}/F_n) . \tag{7}$$

To find the ratio f of the number of orbits oriented to the number of atoms, we may assume that each electronic orbit has a magnetic moment of one Bohr magneton. This ratio then becomes

$$\frac{1}{5589} \times \frac{\text{saturation intensity of magnetization}}{\text{number of atoms per molecule}} \times \frac{\text{molecular weight}}{\text{density}} .$$

On this basis, in the case of magnetite, half of the atoms will have one atom oriented, i.e. $f = 0.5$ while for silicon steel three orbits per two atoms will be turned around, i.e. $f = 1.5$.

The structure factor F_n of the atom for any order may be determined if the structure factor for the first order is known and if the relative intensity reflected in each order is known. Equation (1) could then be used to give the structure factor. The ratio Z/F of the atomic number to the structure factor for the first order does not vary greatly with different atoms. In the cases of sodium and chlorine this ratio is 1.45 and 1.25 respectively. It is thus reasonable to assume the structure factor for the first order of the iron atom to be about 20 and that of the oxygen atom to be about 6.5. The intensities for the first three orders from the 110 planes of iron have been measured, thus permitting direct substitution* in (1) to find the atomic structure factor for orders 1 and 2. In the case of magnetite we may make use of the fact that the experimental values of F, for reflecting planes that are similar and similarly placed, are experimentally found to vary approximately inversely as the order. In computing the results for magnetite it has accordingly been assumed that $F_n = 20/n$ for the iron atom and $6.5/n$ for the oxygen atom.

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* In making this substitution it is necessary to note that the factor K of Eq. (1) includes the trigonometric factor $(1+\cos^2 2\theta)/\sin 2\theta$ (cf. A. H. Compton, reference 2).

For silicon steel these considerations are sufficient for determining $\Delta F_{en}/F_n$; but for magnetite it is necessary to know how a change in atomic structure factor will affect the intensity when the planes are neither similar nor similarly placed. Bragg[4] has shown that the 110 planes may be represented by a single plane having a weight

$$56+112\cos\pi n/4+168\cos\pi n+128\cos\pi n/2,$$

where n is the order of the spectrum. The first three terms are the contributions due to the iron atoms, while the last term represents the contribution of the oxygen atoms. One may assume that both the oxygen and iron atoms are affected by the magnetic field or that either the iron or oxygen atoms have their electronic orbits oriented by the applied field. The first assumption seems the more reasonable and is the one here employed. It is seen that in the fourth order the planes made up of oxygen atoms cooperate with those made up of iron atoms, and a maximum change in intensity would result if only the oxygen atoms were influenced. This is due to the fact that the change in intensity per oxygen atom is greater than that per iron atom and the total number of atoms affected in either case must be the same. If only the iron atoms were influenced the effect would still be detectable, while the result given in the table is that based on the assumption that both iron and oxygen atoms are turned about by an external magnetic field. In the case of the fifth order all the planes may be represented by a plane of iron atoms of weight 90. In the sixth order all the 110 planes may be replaced by planes or iron atoms of weight 224 with planes of oxygen atoms of weight 128 midway between the iron planes. If both kinds of atoms are affected by the magnetic field the change in intensity would be too small to detect, while if the inner orbits of the iron atoms alone are oriented there should be a detectable increase in the intensity of the reflected x-ray beam. On the other hand a change in the oxygen atom alone should produce a decrease in intensity which would have been detected. Thus, by considering the fourth, fifth and sixth orders of magnetite, all possible cases have been considered, and the magnitude of the theoretical effect found is great enough for detection by the method employed whichever atom is affected by the magnetic field. The radii used for the K and L orbits were computed by use of Bohr's formula.

It appears that any reasonable assumptions regarding atomic structure factor or magnetic moment per electron orbit must lead to a predicted effect of the order of magnitude here calculated.

EXPERIMENTAL RESULTS

In the following table both the theoretical and experimental results are given. In the first column on the left the order is given. In the next column is given the theoretical ratio, $I'/I(K)$, of the integrated intensities reflected by the crystal when it is magnetized and unmagnetized respectively, assuming that the K electrons are the ones that orient themselves when the field is applied. In the next column is the same ratio for the case of the L electrons, while the last gives the experimental value.

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[4] W. H. Bragg, Phil. Mag. **30**, 305 (1915).

TABLE I. *Ratio of x-ray reflection from magnetized to that from unmagnetized crystals.*

1. Magnetized perpendicular to the reflecting face			
		Magnetite (111) Plane)	
Order	Calculated I'/I(K)	Calculated I'/I(L)	Experiment I'/I
1	1.0000	1.0000	
2	1.0000	1.0003	1.0001+.0002
3	1.0006	1.0020	0.9999+.0002
4	1.0070	1.0230	0.9998+.0001
5	1.0015	1.0100	1.0002+.0001
6	1.0030	1.0210	1.0001+.0004
7	1.0050	1.0280	0.9988+.0015
		Silicon steel (110 Plane)	
1	1.0000	1.0012	1.0001+.0001
2	1.0009	1.0100	0.9999+.0001
2. Magnetized parallel to the reflecting face			
		Magnetite (111 Plane)	
4	0.9980	0.9100	1.0002+.0010
5	0.9970	0.8900	0.9997+.0004
6	0.9950	0.8000	1.0004+.0003
		Silicon steel (110 Plane)	
2	1.0000	0.9993	1.0003+.0006

In the above table the probable error of the mean is quoted.

It will be seen that the apparent changes in intensity of x-ray reflection due to magnetization are very small, and are of the magnitude to be expected from the probable error. There is thus no evidence for any change whatever in the reflecting power of these magnetic crystals due to magnetization. This conclusion is a considerable extension of the negative result of A. H. Compton and Rognley, for whereas in their experiments magnetite only was used, and this was magnetized only to one third saturation, here both the magnetite and the iron crystals were magnetically saturated. Moreover, these authors gave 1 percent as the precision of the experiment, whereas the more precise of the present experiments may probably be relied upon to 0.1 percent.

SIGNIFICANCE OF THE NEGATIVE RESULTS

Compton and Rognley[1] pointed out that a change of intensity of less than one percent in the fourth order when the crystal was only one third saturated was to be interpreted as meaning that the ultimate magnetic particle could not be the molecule or the atom as a whole unless the atom is surprisingly isotropic. The present data set a lower limit to the maximum displacement of atoms from their normal positions which could have occurred due to magnetization without being detected by this experiment. This distance could not have changed by as much as 1/2000 of the grating space without producing an effect that would have been detected by this experiment. It is also very improbable that the electrons revolving in inner orbits can account for ferromagnetism. In the fourth order in the case of magnetite the theoretical value is 35 times the experimental value, while in the second order for silicon steel the theoretical value is 9 times the experimental change.

Honda[5] ascribes the magnetic property of an atom to the nucleus. Within the nucleus he pictures electrons and protons as revolving in opposite senses. This gives a large magnetic moment and a small angular momentum which would enable the nucleus to be oriented by an applied field. Such a system would be dynamically unstable. Further, as pointed out by Compton and Rognley, the magnetic properties of a substance are vitally affected by temperature, chemical combination and modification of the crystal structure as in the case of the Heusler alloys. On the other hand we cannot, by the most drastic of physical or chemical means, affect the radiation from radioactive substances, thus indicating that the nucleus is not influenced by external conditions. Thus while the results of this experiment are compatable with the theory of Honda, auxiliary evidence seems to indicate that the nucleus cannot be the ultimate magnet.

Ehrenfest has attempted to identify the ultimate magnet with an electron moving within a fixed orbit. In the absence of a magnetic field either sense of revolution is equally probable, while the application of an external magnetic field increases the probability that the sense of revolution of the electron will be such as to produce a magnetic moment in the direction of the field. When one tries to picture the process of magnetization, he is forced to imagine the orbits as turning suddenly, or the electron as suddenly reversing its direction in the orbit. If the whole orbit is able to turn it is difficult to conceive of it as a fixed orbit. If one is to account for hysteresis, energy must be dissipated when the probability of orientation in different directions changes. It then becomes difficult to see how hysteresis can be greatly reduced when the change in direction of magnetization is effected by means of a rotating magnetic field instead of an alternating field.

The Spinning Electron as the Ultimate Magnet

The results of other investigators indicate that neither minute crystal aggregations nor the chemical molecule nor the Rutherford type of atom as a whole can account for ferromagnetism. This experiment shows that it is highly improbable that electrons revolving in any of the orbits are responsible for ferromagnetism. Auxiliary evidence is against the conclusion that the nucleus may account for magnetic properties. It therefore appears that the ultimate magnetic particle should be identified with the spinning electron, as was long ago suggested by Parson[6]; and in accord with Uhlenbeck and Goudsmit's[7] hypothesis of the origin of certain spectroscopic phenomena.

The author wishes to thank Professor A. H. Comptom for proposing the problem, and Dr. R. D. Bennett for constructive suggestions during the course of the experimental work. The kind cooperation of Dr. T. Y. Yensen of the Westinghouse Electric and Manufacturing Company, Professor L. W. McKeehan of Yale University, and Mr. Norman Goss of the American steel and Wire Company in supplying large crystals of iron and silicon steel has made the completion of the work possible.

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[5] Honda, "Theory of Magnetism," p. 192.

[6] A. L. Parson, Smithsonian Misc. Collections, Nov., 1915.

[7] Goudsmit and Uhlenbeck, Nature **117**, 264 (1926).