ON A KINETIC THEORY OF MAGNETISM IN GENERAL.

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1. In the last fifteen years, our knowledge of the nature of magnetism has made a great advance. The electron theory was first proposed by J. J. Thomson[1] and W. Voigt[2]; but their results were not conclusive. P. Langevin[3] developed a kinetic theory of paramagnetism as well as diamagnetism, and arrived at results which agree in many points with the observed facts; but there are many other facts which cannot be explained by his theory. Prof. P. Weiss founded his well-known theory of magnetons[4] on the basis of Langevin's theory of paramagnetic gas; but subsequent investigations[5] showed that the existence of the magneton —the fundamental unit of magnetism—is very doubtful. One of the present writers[6] modified Langevin's theory of the paramagnetism of gases so as to include the magnetization of liquid and solid states, and made clear the distinction between paramagnetic and diamagnetic substances. This theory explains the fact of the dependency of diamagnetic susceptibility on the temperature, and also its change with the change of state; while according to Langevin's theory, such changes of diamagnetism are improbable. Professor Weiss, by introducing a molecular field of enormously large magnitude (some 10^7 gausses) into Langevin's theory of paramagnetic gas, tried to explain the ferromagnetism; but although he succeeded in explaining some of the thermomagnetic properties of ferromagnetic substances, it is very difficult to understand from his theory, why magnetization is so easily induced by a small external field in the presence of the large molecular field. On several occasions, one of the present writers[7] has shown that the existence of the molecular field is very improbable and that the thermomagnetic properties accounted for by means of the molecular field can be equally well explained by other theories.

[1] Phil. Mag., 6, 1903, 673.

[2] Ann. der Phys., 9, 1902, 115.

[3] Ann. de chem. et phy., 5, 1905, 70.

[4] Arch. des Sci., 31, 1911, 401.

[5] K. Honda & T. Ishiwara, Sci. Rep., 4, 1915, 215. K. Honda & H. Takagi, Sci. Rep., 4, 1915, 261; Sci. Rep., 1, 1912, 229.

[6] K. Honda, Sci. Rep., 3, 1914, 171.

[7] K. Honda, Sci. Rep., 4, 1915, 208. K. Honda & J. Okubo, Sci. Rep., 5, 1916, 207. K. Honda, Sci. Rep., 7, 1918, 53.

Various theories of ferromagnetism have been advanced by Poisson, Weber, Maxwell and Ewing. Ewing's theory of molecular magnetism[1] explains the curve of magnetization and the phenomenon of hysteresis quite satisfactorily; but the qualitative nature of the theory is its great defect. R. Gans[2] tried to treat Ewing's model of molecular magnets mathematically; but his theory differs essentially from that of Ewing in assuming the distribution of the molecular magnets in the substance to be quite arbitrary, and in fact the conclusions from his theory are a rough approximation to the observed facts. In the last two years, we have published three papers,[3] in which Ewing's theory of molecular magnetism is treated quantitatively and extended so as to include the theory of hysteresis-loss, as well as the effect of temperature on magnetization. We have obtained the curve of magnetization possessing all the characteristics belonging to iron. Not only has the well-known hysteresis-loop been obtained, but the loss is expressed in terms of the magnetic field and the two constants characteristic of a substance The effect of temperature on the magnetization, that is, the complicated course of magnetization-temperature curves, is also explained on the basis of the same theory.

2. The theory of magnetism in general may thus be considered as established in the main; but there still remains a great advance to be made. As to Langevin's theories of paramagnetic and diamagnetic substances, some modifications are necessary in order to account for the observed facts. For example, take the case of diamagnetic substances: According to Langevin, the diamagnetic susceptibility k per unit of volume of a substance is given by

$$k = -\frac{\rho}{12}\left(\frac{e}{m}\right)^2 r^2,$$

where ρ is the density of electrons, e/m the ratio of the charge to its mass. and r the radius of the orbit of electrons. The susceptibility x per one gram atom or molecule is given by

$$x = -\frac{W\rho}{12D}\left(\frac{e}{m}\right)^2 r^2,$$

where W is the atomic or molecular weight of the substance and D its density. Now, for the density of electrons, we have

$$\rho = \frac{DN}{1800W},$$

[1] Phil. Mag., (5), 30, 1891, 205; Magnetic Induction in Iron and other Metals.

[2] Gött. Nachr., 1910, 197; 1911, 118. R. Gans & P. Hertz, Zeitsch. für Math. u. Phys. 61, 1913, 13.

[3] Sci. Rep., 5, 1916, 153; 5, 1916, 325; 6, 1917, 183.

provided each atom or molecule contains N electrons. Hence for different elements, we have

$$x = -\frac{N}{12 \times 1800}\left(\frac{e}{m}\right)^2 \Sigma r^2,$$

where Σ is to be extended for all electrons. Here we may take N as the atomic number of the elements. Thus, according to Langevin, since Σr^2 is to be considered as increasing with atomic weight, the diamagnetic susceptibility of the elements must increase with their atomic number. But, actually this is not the case; in fact, the susceptibility varies periodically with the atomic weight of the elements. This discrepancy is, however, partially reconciled, if we consider the diamagnetism actually observable to be the differential effect of the paramagnetic and diamagnetic phenomena;[1] that is, denoting the susceptibility observed by x, and Langevin's paramagnetic and diamagnetic susceptibilities by x_p and x_d respectively, we obtain

$$x = x_p + x_d;$$

hence though x_d increases proportionally with the atomic number, x may change periodically, provided x_p makes a similar change.

3. Next, consider the paramagnetic theory of Langevin. According to the kinetic theory of gases, the molecules of a gas are continuously making translational and rotational motions. We suppose each molecule to be a magnet composed of a set of revolving electrons. At a given instant, we may suppose the directions of the axes of these magnetic molecules to be uniformly distributed in all directions, if acted on by no external field. At the next instant, all the magnets change the directions of their axes; but as a whole, these axes are again equally distributed in all directions, and so on. In the case, when an external field acts on them, Langevin considers that the axes of these molecules tend to direct themselves toward the direction of the field; but the thermal impacts prevent this tendency. These two opposing effects make the distribution of the axes of the molecular magnets a little denser in the direction of the field than in the opposite direction. The directions of the magnetic axes are of course changing from instant to instant, but as a whole, the above distribution may be assumed to remain unchanged. By following the analogy of the distribution of the density of a gas under the action of gravity, he assumed the distribution of the axes of the molecular magnets to be given by Maxwell-Boltzmann's law. Resolving their magnetic moments in the direction of the field, and summing up these components, he obtained the well-known expression for the paramagnetic susceptibility.

[1] K. Honda, Sci. Rep., 3, 1914, 171; J. Kunz, PHYS. REV., 6, 1915, 113.

Now, since the molecules are continuously rotating, things are not so simple as Langevin considers. Generally the axis of rotation of a molecule does not coincide with its magnetic axis; hence we consider the magnetic moment to be decomposed into two components parallel and perpendicular to the axis of rotation. The former component is here termed the axial component of the moment and the latter the transverse component. When a magnetic field is suddenly applied to the molecule, in virtue of the axial component of the magnetic moment, its axis of rotation makes a small gyrostatic motion, that is, it makes procession and nutation round the direction of the field. This motion is very little affected by the presence of the transverse component of the moment. The amplitude of the nutational motion measured from the direction of the field extends from an initial angle α to an angle θ towards the field. This motion must therefore make a certain contribution to the increase of the magnetic moment in the direction of the field, while the precessional motion itself does not cause any change in the magnetic moment. If we suppose that by continuous thermal impacts with other molecules, the nutational motion, whose frequency is much larger than that of the precession, rapidly decreases, while the precessional velocity does not considerably alter, a simple precessional motion results with an angle θ, which is greater than the initial angle α and less than the angle θ, and causes also an increase of the magnetic moment in the direction of the field. If the field, instead of being applied suddenly, be gradually applied and increased extremely slowly to its final value, the molecules will make such slow nutational and precessional motions that the first precessional motion at the lower limit may be considered as statical. This case is analogous to that of a pendulum acted on extremely slowly by the force of gravity, in which case, the pendulum may be assumed to remain at rest at its lowest point. If, under the action of a magnetic field, the molecules make a simple precessional motion or take definite orientations, as explained above, it is evident that his case is almost the same as that treated by Langevin. Taking the sum of the magnetic moments in the direction of the field due to all molecules, we obtain usually a very small moment, that is, the paramagnetic moment. By increasing the field, the angular distance $\alpha-\theta$ increases for all molecules, and therefore the paramagnetism of the gas increases with the field. As will be shown later, this result almost coincides with that of Langevin.

4. Consider next the transverse component of the magnetic moment. This component of magnetism is continuously rotating with the molecule in its equatorial plane. Let us at first suppose that this plane coincides with the plane of the magnetic field. Initially its angular velocity is

uniform for all directions during a complete revolution; but if a magnetic field acts on the molecule, the revolution is now not uniform. Toward the direction of the field, the motion is always accelerated; but in the opposite direction, it is always retarded. Hence, if we wish to find its magnetic effect, it is necessary to take the time-mean of the magnetic moment during a complete period of revolution. Since during a revolution, the plus pole of the molecule is a shorter time in the direction of the field than in the opposite direction, the time-mean of the magnetic moment becomes negative, that is, the transverse component of the magnetic moment of rotating a molecule produces a diamagnetic effect. If the equatorial plane of the molecule makes some angle with the plane of the field, it is only necessary to resolve the above effect in the plane of the field. Hence, if there is a number of molecules, the directions of the equatorial planes of which are uniformly distributed between the angles 0 and π, we must take the component moments for these molecules in the direction of the field and integrate them; the result is a diamagnetism. Langevin did not in the least consider the effect corresponding to this kind of diamagnetism. The total magnetic effect of rotating molecules is then the differential effect due to axial and transverse components of magnetic moment, and therefore it may be paramagnetic or diamagnetic, according as the effect due to the axial component is greater or less than that due to the transverse component.

If the above view be correct, Langevin's theory of paramagnetic gas is not valid; and therefore the theory of magnetons, as well as that of molecular field put forward by Prof. Weiss, which are based on the Langevin theory, have no foundation. For this reason, the above view of the magnetization of a gas is very important.

5. We shall next consider the results qualitatively explained above, more in detail. Let us suppose that the magnetic field is acting on a

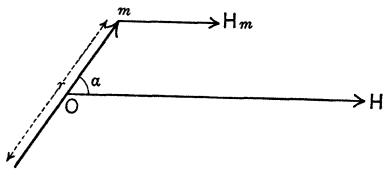

Fig. 1.

molecule of a gas rotating about its magnetic axis; then it will make precessional and nutational motions about the direction of the field. Let o be the center of the magnetic molecule, its pole-strength and pole-distance being m and r respectively. Let α be the initial angle of a molecular magnet, which is acted on by an external field H. Then the

magnetic moment M is

$$M = mr.$$

The following notations are here used:

K = moment of inertia about the magnetic axis,

ω = angular velocity about the magnetic axis,

$\dot{\psi}$ = precessional velocity,

$\dot{\theta}$ = nutational velocity.

From the theory of the gyroscope, we know that

(*i*) the angular momentum about oH remains constant,

(*ii*) the sum of the kinetic and potential energies of the gyro-molecule is constant;

that is,

$$K\omega \cos\theta + K\dot{\psi} \sin^2\theta = K\omega \cos\alpha,$$

$$\tfrac{1}{2}K\omega^2 + \tfrac{1}{2}K\dot{\psi} \sin^2\theta + \tfrac{1}{2}K\dot{\theta}^2 - MH \cos\theta = \tfrac{1}{2}K\omega^2 - MH \cos\alpha;$$

Hence

$$\dot{\psi} = \frac{K\omega(\cos\alpha - \cos\theta)}{K \sin^2\theta}$$

and

$$K\omega^2(\cos\theta - \cos\alpha)^2 + K^2\dot{\theta}^2 \sin^2\theta\, \dot{\theta} = 2MHK\,(\cos\theta - \cos\alpha).$$

The limits of the nutational angle θ and θ_1 are given by

$$\text{Cos}\,\theta = \text{Cos}\,\alpha,$$

$$\text{Cos}\,\theta_1 = -\frac{1}{a} \pm \sqrt{1 + \frac{2}{a}\cos\alpha + \frac{1}{a^2}},$$

where

$$a = \frac{MH}{\tfrac{1}{2}K\omega^2}.$$

In the last equation, $\cos\theta$ must always be less than 1; hence the plus sign must be taken for the double sign. We have then

$$\theta = \alpha,$$

$$\cos\theta_1 = -\frac{1}{a} + \sqrt{1 + \frac{2}{a}\cos\alpha + \frac{1}{a^2}}.$$

Hence the gyro-molecule makes a nutational motion between α and θ_1.

Suppose at first that the magnetic field is applied infinitely slowly, or that the field is suddenly applied, but the nutational motion is assumed to subside very rapidly, the angle θ for the precessional motion being the same as in the above case. This last supposition may not be strictly true, but as we shall see later, if a be very small, the result does not materially differ from that of a case free from the above supposition. In a case, where a is small, the above supposition gives the result, which

exactly coincides with that of Langevin. The magnetic moment in the direction of the field is then $M\cos\theta_1$. The total magnetic moment per one gram molecule is

$$\sigma = \frac{Mn}{2}\int_0^{\pi} \cos\theta_1 \sin\alpha d\alpha$$

$$= \frac{Mn}{2}\int_0^{\pi}\left\{-\frac{1}{a}+\sqrt{1+\frac{2}{a}\cos\alpha+\frac{1}{a^2}}\right\}\sin\alpha d\alpha$$

where n is the number of molecules per one gram molecule of the gas. Writing $Mn = \sigma_0$, we have

$$\frac{\sigma}{\sigma_0} = \frac{1}{2}\int_0^{\pi_0}\left\{\sqrt{1+\frac{2}{a}\cos\alpha+\frac{1}{a^2}}-\frac{1}{a}\right\}\sin\alpha d\alpha$$

$$= \frac{1}{2}\int_{-1}^{+1}\sqrt{1+\frac{1}{a^2}+\frac{2}{a}x}\,dx - \frac{1}{a} = \frac{a}{6}\left|\sqrt{1+\frac{2}{a}x+\frac{1}{a^2}}\right|_{-1}^{+1} - \frac{1}{a}$$

$$= \frac{a}{6}\left|\left\{\left(1+\frac{1}{a}\right)^2\right\}^{\frac{3}{2}} - \left\{\left(1-\frac{1}{a}\right)^2\right\}^{\frac{3}{2}}\right| - \frac{1}{a}.$$

Hence if $a < 1$, we have, taking the minus sign for the square root of $\left(1-\frac{1}{a}\right)^2$,

$$\frac{\sigma}{\sigma_0} = \frac{a}{3}. \tag{1}$$

but if $a > 1$, we get, taking the plus sign for the square root,

$$\frac{\sigma}{\sigma_0} = 1 - \frac{1}{a} + \frac{1}{2a^2}. \tag{1'}$$

Since $\frac{1}{2}K\omega^2$ is the rotational kinetic energy of the molecule, a has the same meaning as that in the Langevin theory, that is,

$$a = \frac{MH}{rT},$$

where r is the gas constant referred to one molecule.

According to Langevin,

$$\frac{\sigma}{\sigma_0} = \operatorname{Coth} a - \frac{1}{a}; \tag{2}$$

if a be small, we have, by expansion,

$$\frac{\sigma}{\sigma_0} = \frac{1}{3}a - \frac{2}{90}a^3 + \cdots \tag{3}$$

which almost coincides with equation (1). In the following table, the two sets of values for σ/σ_0, as calculated from equations (1) and (2), are given side by side:

a	σ/σ_0 (Honda & Ôkubo).	σ/σ_0 (Langevin).	a	σ/σ_0 (Honda & Ôkubo).	σ/σ_0 (Langevin).
0.0	0.0000	0.0000	5.0	0.8133	0.8000
0.5	0.1667	0.1640	6.0	0.8426	0.8334
1.0	0.3333	0.3130	7.0	0.8638	0.8571
2.0	0.5833	0.5373	8.0	0.8802	0.8750
3.0	0.7037	0.6717	9.0	0.8930	0.8889
4.0	0.7708	0.7507	10.0	0.9033	0.9000

From the above table, we see that for small values of a, our formula exactly coincides with that of Langevin; but for a moderate value of a, these two slightly deviate from each other. In Fig. 2, these results (curves 1) are graphically given.

6. Next, we shall consider the case, when the magnetic field is applied

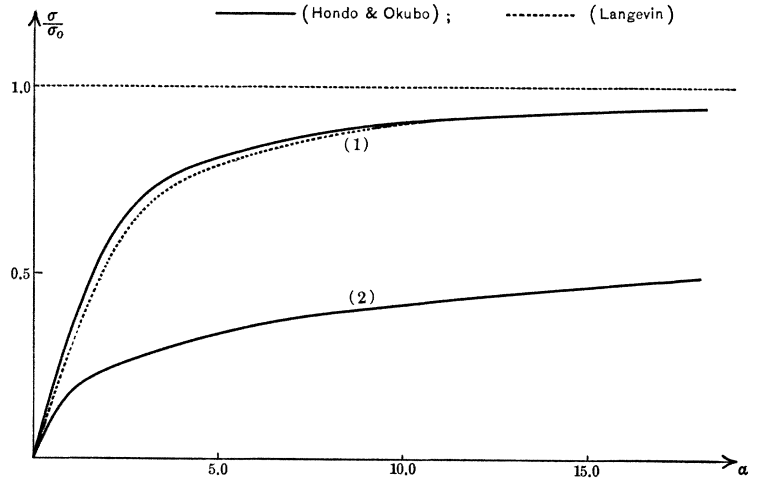

Fig. 2.

impulsively and the consequent nutational motion continues for a long time.

Now, the nutational velocity is given by

$$K \sin^2\theta \left(\frac{d\theta}{dt}\right)^2 = MH (\cos\theta - \cos\alpha)(\cos\theta - \cos\theta_1)(\cos\theta_2 - \cos\theta).$$

Putting

$$\cos\theta = \cos\theta_1 \cos^2\varphi + \cos\alpha \sin^2\varphi$$

and changing the variable from θ to φ, we get

$$\left(\frac{d\varphi}{dt}\right)^2 = \frac{MH}{4K}(\cos\theta_1 + \cos\theta_2)\,(1 - \kappa^2\sin^2\varphi),$$

where

$$\kappa^2 = \frac{\cos\theta_1 - \cos\alpha}{\cos\theta_1 + \cos\theta_2}.$$

Hence

$$\frac{dt}{p} = \frac{d\varphi}{\sqrt{1 - \kappa^2\sin^2\varphi}},$$

where

$$p = \sqrt{\frac{MH}{4K}(\cos\theta_1 + \cos\theta_2)}.$$

Let us suppose that

$$\text{at } t = T, \qquad \theta = \alpha, \qquad \varphi = \frac{\pi}{2},$$

and

$$\text{at } t = 0, \qquad \theta = \theta_1, \qquad \varphi = 0,$$

then

$$\frac{T}{p} = \int_0^{\pi/2} \frac{d\varphi}{\sqrt{1 - \kappa^2\sin^2\varphi}} = K(\kappa),$$

where $K(\kappa)$ is the complete elliptic integral of the first kind.
θ or φ at any instant is given by

$$\frac{t}{p} = \int_0^{\phi} \frac{d\varphi}{\sqrt{1 - \kappa^2\sin^2\varphi}} = F(\varphi, \kappa).$$

$F(\varphi, \kappa)$ is the Legendre elliptic integral of the first kind, that is,

$$\varphi = am\frac{t}{p}$$

and

$$\cos\theta = \cos\theta_1\, cn^2\frac{t}{p} + \cos\alpha\, sn^2\frac{t}{p}.$$

Hence the time-mean of the magnetic moment is

$$\begin{aligned} M_m &= \frac{1}{T}\int_0^T M\cos\theta\, dt \\ &= \frac{M}{T}\left\{\cos\theta_1\int_0^T cn^2\frac{t}{p}\,dt + \cos\alpha\int_0^T sn^2\frac{t}{p}\,dt\right\} \\ &= \frac{Mp}{T}\left|\cos\theta_1\left\{\frac{1}{\kappa^2}\left[E\left(am\frac{t}{p},\ \kappa\right) - \kappa'^2\frac{t}{p}\right]\right\}\right. \\ &\qquad \left. + \cos\alpha\left\{\frac{1}{\kappa^2}\left[\frac{t}{p} - E\left(am\frac{t}{p},\ \kappa\right)\right]\right\}\right|_0^T. \end{aligned}$$

But since

$$\frac{p}{T} = K(\kappa) \text{ and } amK = \frac{\pi}{2},$$

$$\therefore \frac{M_m}{M} = \frac{\cos\theta_1 - \cos\alpha}{\kappa^2}\frac{E(\kappa)}{K(\kappa)} + \frac{1}{\kappa^2}(\cos\alpha - \kappa'^2\cos\theta_1)$$

$$= (\cos\theta_1 + \cos\theta_2)\frac{E(\kappa)}{K(\kappa)} - \cos\theta_2.$$

If the axis of the molecular magnets are initially distributed uniformly in all directions, the resultant magnetic moment in the direction of the field is

$$\frac{\sigma}{\sigma_0} = \frac{1}{2}\int^{\pi}\left\{(\cos\theta_1 + \cos\theta_2)\frac{E(\kappa)}{K(\kappa)} - \cos\theta_2\right\}\sin\alpha d\alpha. \tag{4}$$

Since $\cos\theta_1$ and $\cos\theta_2$ are all functions of α, the above integral cannot be evaluated in a rational form. If a be small, $\cos\theta_1$, κ, $E(\kappa)$, $K(\kappa)$ can be expanded in the ascending power series of a and then integrated, we have

$$\frac{\sigma}{\sigma_0} = \frac{1}{3}a - 0.0334a^3 + \cdots. \tag{5}$$

In a case when a is very large, these quantities can be expanded in an ascending power series of $\lambda = 1/a$; then we get

$$\frac{\sigma}{\sigma_0} = 0.4818 - 0.6875\lambda + 0.6476\lambda^2 + \cdots. \tag{6}$$

This equation shows that even in an infinitely large field, the magnetic moment does not attain its saturation value; because for $H = \infty$, $\lambda = 0$. Hence

$$\frac{\sigma}{\sigma_0} = 0.4818.$$

The reason for this is evident, since according to our first supposition, all the molecules are making continuously nutational oscillations like a pendulum about the direction of the field.

By using equations (5) and (6) for small and large values of a, and also by mechanical integration of equation (4) for moderate values of a, we calculated the values of σ/σ_0 and the result is given in Fig. 2, curve 2. In ordinary cases actually realizable, in which a is a very small quantity, equations (3) and (5) coincide with each other; but as the values of a becomes large, the deviation between them becomes always greater. To decide which one of these equations for a large value of a actually corresponds to the observed facts, requires a further experimental investigation.

7. Next, we shall consider the case of a molecule rotating about an axis perpendicular to the magnetic axis. If the plane of rotation coincides with that of the field, the time-mean of the magnetic moment M_m in the direction of the field[1] is given by

$$\frac{M_m}{M} = \frac{2}{\kappa^2}\frac{E(\kappa)}{K(\kappa)} - \left(2\frac{\kappa'^2}{\kappa} + 1\right),$$

where

$$\kappa^2 = \frac{4MH}{K\omega^2} = 2a.$$

If the plane of rotation makes an angle θ with the direction of the field, its effective moment is $M_m \cos\theta$. Hence if the distribution of the planes of rotation for n molecules be uniform, the resultant moment in the direction of the field will be

$$\sigma = \frac{M_m n}{2}\int_0^\pi \cos\theta \sin\theta d\theta = \frac{M_m n}{2},$$

or

$$\frac{\sigma}{\sigma_0} = \frac{1}{2}\left\{\frac{2}{\kappa^2}\frac{E(\kappa)}{K(\kappa)} - \left(2\frac{\kappa'^2}{\kappa^2} + 1\right)\right\}. \tag{7}$$

If a or κ be small, we have by expansion

$$\frac{\sigma}{\sigma_0} = -\frac{a}{8} - \frac{a^2}{8} - \cdots. \tag{8}$$

This shows that the molecules rotating about the axis perpendicular to the magnetic axes produce the diamagnetism. Since $\frac{1}{2}K\omega^2$ in the expression for a is the rotational energy of the molecules, its value is of the same order of magnitude as rT. Hence even in the strongest field available at the present day, the value of a is very small, unless the temperature is very low. Hence we may safely neglect a^2 in comparison with a, and we obtain

$$\frac{\sigma}{\sigma_0} = -\frac{a}{8}. \tag{9}$$

8. Having thus found the magnetic effects of the gas molecules for the two cases above mentioned, we now proceed to consider the case of the magnetization of an actual gas. Since the molecules are always rotating about axes whose directions are continuously changing, the axis of rotation and the magnetic axis do not generally coincide with each other. Now, at any given instant, among n molecules, there is a certain number of molecules

$$dn = \frac{n}{2}\sin\varphi d\varphi,$$

[1] K. Honda & J. Okubo, Sci. Rep., 5, 1916, 328.

whose magnetic axis make with the axis of rotation an angle lying between φ and $\varphi + d\varphi$. Resolving the magnetic moment of these molecules in the axial and transverse directions, we get $M \cos \varphi$ and $M \sin \varphi$ for these two components respectively. On the one hand, these molecules have then the magnetic moment $M \cos \varphi$ in the direction of the axis of rotation, and therefore by applying a magnetizing field, their magnetic polarization is paramagnetic. Its intensity of magnetization $d\sigma$ is obtained from equation (1), since the quantity corresponding to a is very small; that is,

$$a' = \frac{MH \cos \varphi}{\frac{1}{2}K\omega^2} = a \cos \varphi$$

and

$$d\sigma = M \cos \varphi dn \times \frac{a'}{3} = \frac{Mna}{6} \cos^2 \varphi \sin \varphi d\varphi.$$

φ may take any value between 0 and π; hence integrating this expression for φ, we get the total effect, that is,

$$\frac{\sigma}{\sigma_0} = \frac{a}{6} \int_0^{\pi} \cos^2 \varphi \sin \varphi d\varphi = \frac{a}{9}. \tag{10}$$

On the other hand, the magnetic effect due to the transverse component of magnetic moment for the molecules is obtained from equation (9). Thus,

$$a' = \frac{HM \sin \varphi}{\frac{1}{2}K\omega^2} = a \sin \varphi$$

and

$$d\sigma = - M \sin \varphi dn \times \frac{a'}{8}$$

$$= - \frac{Mna}{16} \sin^3 \varphi d\varphi.$$

Integrating this expression for φ between 0 and π, we get the total effect, that is,

$$\frac{\sigma}{\sigma_0} = - \frac{a}{16} \int_0^{\pi} \sin^3 \varphi d\varphi = - \frac{a}{12}. \tag{11}$$

Hence, if the principal moments of inertia are equal to each other, that is, the shape of the molecules is spherical, the resultant moment of n molecules is the sum of the expressions (10) and (11). That is,

$$\frac{\sigma}{\sigma_0} = \frac{a}{3} (\tfrac{1}{3} - \tfrac{1}{4}) = 0.0278a. \tag{12}$$

Thus the resultant is paramagnetic. This differs quantitatively from the result of Langevin.

It is commonly assumed that the molecules of a monoatomic gas have a spherical shape and therefore thermal impacts cannot produce any rotation of the molecules and they have no energy of rotation. But according to a theory[1] proposed by one of the present writers, the molecules of the monoatomic gas are also continuously rotating with great velocity, which is acquired in a liquid state, but this velocity does not depend on the temperature of the gas. Hence, even for the monoatomic gas, the above result is equally applicable.

If the form of the molecules is a body of revolution about the magnetic axis, we have

$$a = \frac{MHK'}{\frac{1}{2}K^2\omega^2} = \frac{\sigma_0 H}{q}\frac{K'}{K}$$

and

$$a' = \frac{MH}{\frac{1}{2}K'\omega'^2} = \frac{\sigma_0 H}{q},$$

where K and K' are respectively the moments of inertia about the magnetic axis and the axis perpendicular to it, and

$$q = \frac{n}{2}K\omega^2 = \frac{n}{2}K'\omega'^2$$

is the kinetic energy of rotation, the equipartition of the rotational energy being assumed. Hence,

$$\frac{\sigma}{\sigma_0} = \left(\frac{K'}{3K} - \frac{1}{4}\right)\frac{\sigma_0 H}{3q}. \tag{13}$$

The gas may therefore be paramagnetic or diamagnetic, according as

$$4K' - 3K \gtrless 0.$$

As an example, take an ellipsoidal molecule, whose form is defined by

$$\frac{x^2}{A^2} + \frac{y^2 + z^2}{B^2} = 1$$

and whose major axis x coincides with that of its magnetic axis; then we have

$$K = \frac{M}{5} \times 2B^2 \text{ and } K' = \frac{M}{5}(A^2 + B^2).$$

Hence the gas is paramagnetic or diamagnetic according as

$$4A^2 - 2B^2 \gtrless 0,$$

or

$$\frac{A}{B} \gtrless 0.7072,$$

or

$$\text{eccentricity} \gtrless 0.707.$$

[1] K. Honda, Sci. Rep., 7, 1918.

It is an interesting fact that the sign of the magnetization of a gas depends only on the shape of the molecules, and not in the least on their the magnetic moment.

9. It is generally admitted that the molecular magnets consist of certain sets of electrons rapidly revolving about positive nuclei, and that this revolution of electrons does not vary with temperature, its velocity always remaining the same, even at absolute zero. This rotational energy may be called the zero point energy; we denote it by $\frac{1}{2}R\Omega$, where R is the gas constant referred to one gram molecule. Besides, at the temperature T, we have also the axial component of rotation of the molecules as a whole; its energy may be given by $\frac{1}{2}RT$. We have then

$$\frac{n}{2}K\omega^2 = \tfrac{1}{2}RT + \tfrac{1}{2}R\Omega = \tfrac{1}{2}R(T+\Omega).$$

Hence equation (13) becomes

$$\frac{\sigma}{\sigma_0} = p\frac{\sigma_0 H}{R(T+\Omega)},$$

where

$$p = \frac{2}{3}\left(\frac{K'}{3K} - \frac{1}{4}\right);$$

p may be positive or negative; or

$$x = \frac{\sigma}{H} = \frac{p\sigma_0^2}{R(T+\Omega)}. \tag{14}$$

Thus, the paramagnetic or diamagnetic susceptibility is proportional to the magnetic field. From the last equation, we have

$$x(T+\Omega) = \frac{p\sigma_0^2}{R} = \text{const.}$$

If $p>0$ and Ω is very small as compared with T, we have

$$xT = \text{const.},$$

which is the Curie law for paramagnetic gas. On the other hand, if $p<0$ and Ω is very large, we have

$$x = \text{const.},$$

Hence for a gas satisfying these conditions, its diamagnetic susceptibility is independent of temperature.

Here the following remarks are to be made. So far we have assumed the molecular magnets to be perfectly rigid; but as shown by Langevin, the motion of the electrons is slightly modified by applying a strong magnetic field, and this change also contributes to the diamagnetism of

the substance. Hence to equation (14), we must add the diamagnetic susceptibility as given by Langevin, that is,

$$x = -\frac{N}{12 \times 1800}\left(\frac{e}{m}\right)^2 \Sigma r^2.$$

As an application of the above theory, it is interesting to calculate the magnetic moment of a hydrogen atom or molecule. According to Bohr, the atom of hydrogen consists of an electron revolving about its nucleus. Since Bohr's value for the smallest radial distance of the revolving electron seems to be too small, we take its next value for r and the corresponding angular velocity, that is,

$$r = 2.2 \times 10^{-8} \text{ cm.}, \qquad \omega = 0.755 \times 10^{15} \text{ per sec.}$$

Then the magnetic moment of an atom is

$$M = \tfrac{1}{2}er^2\omega = 2.95 \times 10^{-21}$$

and

$$\sigma_0 = 1806,$$

which is a little greater than half the magnetic moment of the nickel atom.

The zero point energy E_0 of rotation amounts to

$$E_0 = 6.2 \times 10^{11};$$

but at 27° C.,

$$\tfrac{1}{2}RT = 1.24 \times 10^{10},$$

which is only a small fraction of the zero point energy. Hence, we have

$$\frac{n}{2}K\omega^2 = E_0 + \tfrac{1}{2}RT = 0.63 \times 10^{11}.$$

In the well known Bohr model of a hydrogen molecule, the two electrons are revolving about the line joining two positive nuclei. Here the moment of inertia K about the magnetic axis is very small as compared with the moment of inertia K' about an axis perpendicular to it; hence the ratio K'/K is very large and therefore the susceptibility of hydrogen must be paramagnetic. This inference, however, contradicts the observed fact. In order therefore that the hydrogen molecule may give the observed diamagnetic susceptibility, the positive nuclei must approach very near to each other.

Let us next calculate the distance d between two positive nuclei of a hydrogen molecule composed of two atoms, the planes of the electron orbits being parallel to each other, under the condition that the susceptibility of the gas may give the value[1] actually observed, that is,

$$x = -1.89 \times 10^{-6}.$$

[1] The measurement of the susceptibility of different gases is now being made in this laboratory by Mr. J. Sonê.

In this case, Langevin's diamagnetic susceptibility amounts to

$$x = -7.0 \times 10^{-6}.$$

Hence by equation (13), we have the relation

$$-1.8 \times 10^{-6} = \frac{1}{3}\left(\frac{K'}{3K} - \frac{1}{4}\right)\frac{\sigma_0^2}{q} + xd;$$

or

$$\frac{K'}{3K} = \frac{1}{12} + 5.4 \times 10^{-6}\frac{q}{\sigma_0^2} = 1.17.$$

$$\therefore \quad \frac{r}{d} = 13.12.$$

Hence the two positive nuclei should be only distant from each other by an amount less than one thirteenth of the radius of the electron orbits.

10. The case of solid substances will be next considered. According to the prevailing theory of the solid state, the molecules are arranged according to a certain space-lattice and are continuously making rectilinear oscillations, but not any rotations. But as we have already remarked, the electrons in each atom are rapidly revolving about the centers of mass of the electrons and the positive nuclei; its axis of rotation coincides evidently with that of the magnetic moment. The energy of this rotational motion, that is, the zero point energy, may be expressed by $\frac{1}{2}\epsilon R\Omega$, where Ω is a constant and ϵ a small fraction of unity. With the rise of temperature, the angular velocity of this rotation may probably increase, because of the axial rotation of the molecules as a whole due to thermal impacts. In a solid, this increase of rotation is not independent of the rectilinear vibrations and therefore the increase of energy is far less than that corresponding to the energy of one independent freedom, that is, than $\frac{1}{2}RT$; hence it may be expressed by $\frac{1}{2}\epsilon RT$. For the molecules of a solid, a simultaneous rotation of the molecules perpendicular to the above rotation can not possibly exist.[1] Hence assuming the molecules to form a numerous number of minute groups, in each of which the axis of magnetic molecules take the same direction, but the magnetic axes of these groups are uniformly distributed in all directions, the expression for the susceptibility of a solid substance is given by equation (1). Since

$$\frac{n}{2}K\omega^2 = \frac{\epsilon}{2}R(T + \Omega),$$

$$\frac{\sigma}{\sigma_0} = \frac{a}{3} = \frac{2}{3}\frac{\sigma_0 K'H}{KR\epsilon(T + \Omega)};$$

[1] K. Honda, Sci. Rep., 7, 1918.

$$x = \frac{\sigma}{H} = \frac{2}{3}\frac{\sigma_0^2 K'}{KR\epsilon(T+\Omega)}; \tag{15}$$

$$\therefore \quad x(T+\Omega) = \frac{2}{3}\frac{\sigma_0^2 K'}{KR\epsilon} = \text{const.}$$

There is a considerable number of paramagnetic substances, whose susceptibility satisfies the above relation. Strictly speaking, to the above value of susceptibility, we must also add the small diamagnetic susceptibility given by Langevin.

According to the theory of fusion[1] proposed by one of the present writers, the rotational motion of the molecules during fusion begins to take place with the energy corresponding to the degree of freedom at the melting point, so that the liquid molecules are making axial and transverse rotations like those of a gas. Hence during fusion, the susceptibility of a substance discontinuously changes from a value expressed by equation (15) to that expressed by equation (14), and may in some cases change its sign. The remarkable fact[2] that during melting, the susceptibility of tin changes from paramagnetic susceptibility to the diamagnetic, is explained in this way.

11. The case of a ferromagnetic substance comes next for our consideration. According to our theory of magnetism,[3] which explains the observed fact in a very satisfactory manner, the molecules of a ferromagnetic substance are arranged in a definite space-lattice and exert mutual magnetic action upon one another. In the light of the present theory, this means that the energy of rotation about the magnetic axis, that is $\frac{1}{2}K\omega^2$, is very small; for in this case, the gyrostatic action of the molecules is negligably small in comparison with the mutual magnetic action, and the molecules are now in equilibrium under the action of this magnetic force, a condition required in our theory.

Now, take equations (1) and (1′) as our starting point; it appears at first sight that the expression

$$\frac{\sigma}{\sigma_0} = \frac{a}{3} \text{ for } a < 1,$$

$$\frac{\sigma}{\sigma_0} = 1 - \frac{1}{a} + \frac{1}{2a^2} \text{ for } a > 1,$$

where

$$a = \frac{\sigma_0^2 HK'}{\frac{n}{2}K^2\omega^2},$$

[1] K. Honda, Sci. Rep., 7, l. c.

[2] K. Honda, Sci. Rep., 1, 1911, 1.

[3] K. Honda and J. Okubo, Sci. Rep., 5, *l. c.*

may be looked on as the law of magnetization for a ferromagnetic substance, provided we consider σ to be a function of H and $(n/2)K\omega^2$ to be a very small quantity depending on the temperature of the substance. The form of the $\sigma_1 H$ curve is the same as curve 1 in Fig. 2. The curve possesses two characteristic points of the curve of magnetization, that is, the proportionality between the initial magnetization and the field, and the gradual approach of magnetization to saturation; yet it is only a rough representation of the fact; moreover it cannot explain the important phenomenon of hysteresis. These discrepancies are the consequences of the present theory, which neglects the mutual magnetic action and takes only the axial rotation of the molecules into account. As we have shown in our former papers, in the case of ferromagnetic substances, we must consider the mutual magnetic action as the most important factor in determining the law of magnetization, and therefore equations (1) and (1'), which take only the gyrostatic action of the axially rotating molecules into account, cannot be considered as the true law of magnetization.

The effect of temperature on magnetization will be next considered. The temperature affects magnetization in two opposite ways; the first effect, which is especially conspicuous in weak fields, but which completely vanishes in strong fields, is to increase the magnetization, while the second is always to decrease the magnetization. In our former paper,[1] these two effects are explained on the basis of small vibratory motions of the molecular magnets, about their mean orientations. That the magnetization in weak fields is considerably increased by the thermal vibrations of the molecules, is very satisfactorily explained; but the explanation of the second effect by the same vibrations is met with the great difficulty that if the half-amplitude of this vibration exceeds about 131°, the magnetization in a strong field is reversed; this, however, is an abnormal result, which has never been observed.

In view of the present theory, the following explanation for the temperature-effect in question will be very natural and agrees satisfactorily with the observed facts: The molecules of a ferromagnetic substance are continually making axial rotations about their magnetic axis, and at the same time, small vibratory motions about the axes perpendicular to it. The first motion explains the diminution of magnetization, while the second motion explains its increase in weak fields, as shown in our former paper. At ordinary temperature, the energy of the axial rotation is considered to be very small, but as the temperature rises, it rapidly increases, its rate of increase becoming always greater. At the critical

[1] Sci. Rep., 5, 1916, 325.

point, the energy is considered to be greatly increased, but it has still a small fraction of the value $\frac{1}{2}RT$ corresponding to one degree of freedom at the temperature T. The energy-content of the axial rotation is then given by an expression of the form

$$\frac{n}{2}K\omega^2 = \frac{m}{2}RT,$$

where m is a function of temperature increasing rapidly with the latter, the zero point energy being neglected. An expression m of the form

$$m = \frac{\epsilon}{\sqrt{1 - \frac{T}{\theta} + \epsilon}},$$

where θ is the critical temperature; e and e_1 being very small quantities, will accord with the observed facts very satisfactorily. The law of variation of magnetization with temperature is then given by equations (1), (1′) and

$$a = \frac{\sigma_0^2 HK'_1}{\frac{1}{2}KmRT}.$$

In order to show that these expressions are a good representation of the

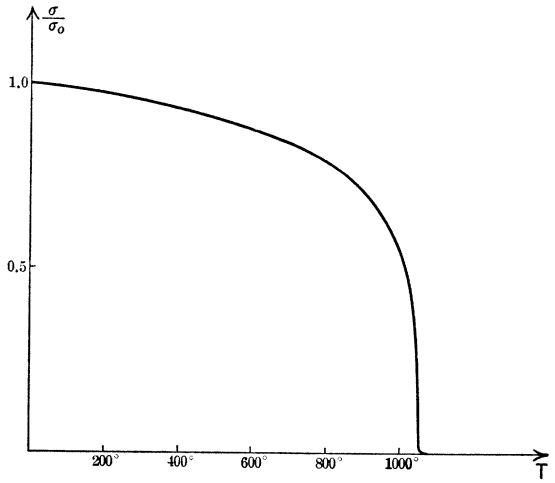

Fig. 3.

observed facts, we have calculated the constants entering into the expression of a for iron and obtained the following results:

$$\theta = 790° \text{ C.},$$

$$\epsilon = 0.431 \times 10^{-5},$$

$$\epsilon_1 = 4.96 \times 10^{-8}.$$

The curve is also given in Fig. 3; we see that it is very similar to the I_1T curve in a strong field. Hence we conclude that the effect of temperature, which diminishes the magnetization for all fields, is due to the increase of the axial rotational energy of the molecules. This inference is also confirmed by the fact that during heating or cooling through the critical range of magnetic transformation, the heat is absorbed or evolved, that is, through the same range, the specific heat abnormally changes.

If the above view be correct, the rotational energy gained during heating through the range of magnetic transformation up to the critical point, is equal to the total heat absorbed during the transformation. If we neglect a small rotational energy at room temperature, the total energy q gained is equal to $(n/2)K\omega^2$. Hence, for one gram-molecule of a ferromagnetic substance, we have

$$x = \frac{2}{3}\frac{K'\sigma_0^2}{nK^2\omega^2} \text{ or } = \frac{K'\sigma_0^2}{3Kq};$$

$$q = \frac{K'\sigma_0^2}{3KX}.$$

This relation admits of the calculation of the heat absorbed or evolved during heating or cooling, provided the shape of the molecules be known. But at present, we have no data for determining the ratio K'/K for ferromagnetic substances, and therefore the verification of the above relation is left for a future occasion.

The following remark deserves also to be made:—The magnetic (or A_2) transformation of iron has been the subject of a long dispute among metallurgists, viz. as to the nature of this transformation.[1] The present theory gives, however, an exact mechanism of this transformation; that is, the A_2 transformation is not a change of molecular configuration or a change in the molecules, as is the case in an allotropic change; but it is the process of acquiring the rotational energy of the molecules, whose amount is a definite function of temperature. Hence the transformation cannot be considered as an allotropic change, as has repeatedly been stated by one of the present writers.[1]

10. Lastly let us consider the susceptibility of a ferromagnetic substance above its critical point. In this region, the kinetic energy of axial rotation, that is, $(n/2)K\omega^2$, is very large in comparison with that in the magnetic region, but it is still far less than the energy corresponding to one degree of freedom at the temperature under consideration, that is, $\frac{1}{2}RT$, which value is only attained by the fusion of the substance. We may express the above fact by the expression

$$\tfrac{1}{2}K\omega^2 = \tfrac{1}{2}RT - \tfrac{1}{2}R\Omega$$

[1] K. Honda, Sci. Rep., 4, 1915, 169; 6, 1917, 213.

and therefore equation (1) becomes

$$\frac{\sigma}{\sigma_0} = \tfrac{1}{3}a = \frac{2\sigma_0 HK'}{KR(T-\Omega)},$$

or

$$x(T-\Omega) = \frac{2\sigma_0^2 K'}{RK} = \text{const.} \qquad (16)$$

This is the relation, which holds good approximately above the critical point. This relation was first obtained by Prof. P. Weiss from his theory of molecular field and given in his paper as a confirmation of his theory. But as was shown by one of the present writers, it can also be obtained from another theory,[1] which has no connection with the molecular field. The above deduction of the same relation affords also a similar example.

The relation (16) may also be written as

$$I(T-\Omega) = \text{const.} \times H.$$

This represents a hyperbola, whose parameter is H; curves 1, 2, 3, in Fig. 4 are those corresponding to successively increasing fields. Curie,

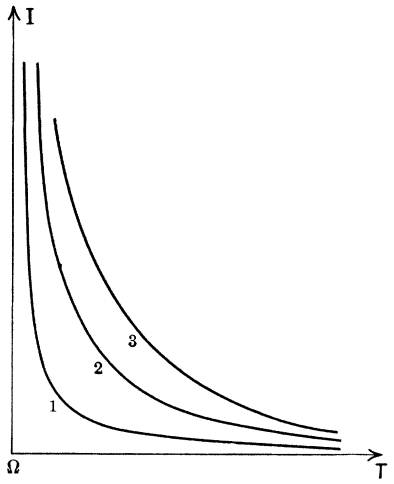

Fig. 4.

in his experimental research on the susceptibility of iron at high temperatures, first noticed this result and remarked that the course of the curves resembles the p, v curve of a gas near its critical point. In recent years, this remark has attracted the attention of many physicists[1]; they have attempted to explain the relation between ferromagnetic and paramagnetic states by the analogy of the relation of the liquid state to the gaseous. But as we have shown above, the fact is a natural consequence from the present theory, and we think it has not the important significance which other physicists assign to it.

[1] P. Weiss & P. N. Beck, Jour. de Phys., 7, 1908, 249. Ashworth, Phil. Mag., 27, 1915, 357; 30, 1915, 711; 33, 1917, 334. E. Bruins, Phil. Mag., 34, 1917, 380.