FERROMAGNETIC RESONANCE ABSORPTION
IN BaFe$_{12}$O$_{19}$, A HIGHLY ANISOTROPIC CRYSTAL *)

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Summary
The ferromagnetic resonance absorption has been measured at 24 000 Mc/s for a single crystal of hexagonal BaFe$_{12}$O$_{19}$. The resonance conditions are severely influenced by the crystalline anisotropy and, for fields too small for saturation, also by the Weiss-domain structure. The theory predicts, for a varying magnetic field perpendicular to the hexagonal axis, at most three absorption peaks, which have been observed at elevated temperatures. The gyroscopic splitting factor $g$ and the anisotropy field are evaluated. The $g$-factor has the spin-only value. The crystalline anisotropy is suggested to be caused by dipole-dipole interaction.

Résumé
L'absorption de résonance ferromagnétique d'un monocristal hexagonal du BaFe$_{12}$O$_{19}$ a été mesurée à 24 000 MHz. Les conditions de résonance sont influencées très gravement par l'anisotropie cristalline et si le champ magnétique est trop petit pour la saturation, elles le sont aussi par les domaines de Weiss. La théorie prévoit au plus trois maxima d'absorption pour un champ magnétique variable perpendiculaire à l'axe hexagonal, maxima qui ont été observés aux températures élevées. Le facteur de séparation gyroscopique $g$ et le champ d'anisotropie sont évalués. Ce facteur $g$ a la valeur du spin seul. Il est probable que l'anisotropie est causée par l'interaction magnétique entre les dipôles.

Zusammenfassung
Die ferromagnetische Resonanzabsorption wurde bei 24 000 MHz an einem Einkristall des hexagonalen BaFe$_{12}$O$_{19}$ gemessen. Die Resonanzbedingungen werden wesentlich beeinflußt von der Kristallanisotropie und — sofern die Felder zur Erzielung einer Sättigung zu niedrig sind — auch von den Weissschen Bezirken. Die Theorie liefert höchstens drei Absorptionsmaxima für ein variabeles Magnetfeld senkrecht zur hexagonalen Achse, welche bei hohen Temperaturen beobachtet wurden. Der gyroskopische Spaltungs faktor $g$ und das Anisotropiefeld sind ausgewertet worden. Der $g$-Faktor hat den nur-Spin Wert. Die Kristallanisotropie wird wahrscheinlich von der magnetischen Dipolwechselwirkung verursacht.

1. Introduction
A ferromagnetic substance, when exposed to a microwave field whose magnetic vector is perpendicular to a static magnetic field $H$, shows resonance phenomena if the frequency is equal to the Larmor frequency, which in an isotropic crystal equals

$$\omega_r = 2\pi f_r = \gamma H = \frac{e}{2mc} gH,$$

(1)

*) A summary of this paper was given by the first author at the Conference on Ferrimagnetism, at the Naval Ordnance Laboratory, Maryland, U.S.A., 11-12 Oct. 1954.
where \( \gamma \) is the gyromagnetic ratio and \( g \) the spectroscopic splitting factor; the latter is equal to 2 for electron spins. This means that the resonance frequency is proportional to the stiffness with which the magnetic moment is bound to the equilibrium orientation. If the crystal is magnetically anisotropic, so that the energy depends upon the orientation of the magnetization with respect to the crystallographic axes, an extra stiffness will be added, which may be either positive or negative. In the cases so far studied this extra anisotropy stiffness was small compared to that due to the external magnetic field at microwave frequencies. This is not the case for the hexagonal crystal \( \text{BaFe}_{12}\text{O}_{19} \), which has a very high crystalline anisotropy \(^1\). The anisotropy energy can in this case be described with the formula

\[
E_c = K \sin^2 \theta, \tag{2}
\]

in which \( \theta \) is the angle between the magnetization vector and the hexagonal axis. This axis is the easy direction of magnetization, so that \( K > 0 \); at room temperature \( K \approx 3 \times 10^6 \text{ erg/cm}^3 \). The stiffness in the easy direction is equal to that of a magnetic field of magnitude \( H^d = 2 K/M \), in which \( M \) is the saturation magnetization; this field has a value of about 17000 oersteds, which corresponds to a wavelength of 6 mm. We shall prove, however, that the resonance frequency can become vanishingly small by the application of a magnetic field in a difficult direction of magnetization which is here perpendicular to the hexagonal axis of the crystal.

Both in the easy and the difficult orientations of the magnetization the energy has an extreme value. This remains so for the difficult direction if we apply a magnetic field in this direction. For small fields it is a maximum, but for strong fields, when the magnetization vector is turned over into this direction, apparently it is a minimum. Consequently for a field strength at which the magnetization vector has just arrived in this orientation the stiffness in the direction from which it arrives is zero. The stiffness for movements in the perpendicular direction can remain finite, but this does not prevent the resonance frequency from becoming zero for this particular value of the field strength. At higher fields \( \omega_r \) increases with \( H \). Thus, for a frequency which is not high enough to excite ferromagnetic resonance with the magnetization vector in the easy direction, one should expect to find resonance at two values of a field in the difficult direction. From these values one can deduce both the \( g \)-factor and the anisotropy field \( H^d \). A more detailed theory will be given in section 2.

Such resonance experiments were carried out on a \( \text{BaFe}_{12}\text{O}_{19} \) single crystal but the situation is there more complicated owing to the fact that a field perpendicular to the easy direction leaves the Weiss-domain pattern undisturbed. This is always the case if there are several easy orientations
which make equal angles with the applied field in the difficult direction. This applies to cubic crystals as well, where only the 180° walls disappear. It will be shown in section 3 that the domain structure changes the resonance conditions completely for a hexagonal crystal.

The experimental technique will be treated in section 4 and the results of the measurements will be given in section 5.

2. Theory of ferromagnetic resonance conditions

The magnetic free enthalpy \( G(T, H) \) of 1 cm³ of a magnetized crystal is, for constant \( T \) and \( H \), a function of the orientation of the magnetization vector. This energy includes that of the static magnetic field, crystalline anisotropy, demagnetization energy, etc. Let the equilibrium direction of the magnetization vector be the \( \zeta \)-direction, and the small angles of deviation in two perpendicular directions \( \xi \) and \( \eta \). The equations of motion are then

\[
\begin{align*}
-M\dot{\eta} &= \gamma \frac{\partial G}{\partial \xi}, \\
M\dot{\xi} &= \gamma \frac{\partial G}{\partial \eta}.
\end{align*}
\]

For small deviations from the equilibrium position we may use for \( G \) the first terms of a Taylor series:

\[
G = G_0 + \frac{1}{2} \left( G_{\xi \xi} \xi^2 + 2G_{\xi \eta} \xi \eta + G_{\eta \eta} \eta^2 \right),
\]

so that (3) becomes

\[
\begin{align*}
-M\dot{\eta} &= \gamma \left( G_{\xi \xi} \xi + G_{\xi \eta} \eta \right), \\
M\dot{\xi} &= \gamma \left( G_{\eta \xi} \xi + G_{\eta \eta} \eta \right).
\end{align*}
\]

This pair of equations has solutions which vary harmonically in time if the angular frequency \( \omega \) satisfies

\[
\omega_r = (\gamma/M) \left( G_{\xi \xi} G_{\eta \eta} - G_{\xi \eta}^2 \right)^{1/2}.
\]

For an arbitrary orientation of the equilibrium position with respect to a coordinate system with polar angles \( (\vartheta, \varphi) \) one obtains

\[
\omega_r = (\gamma/M \sin \vartheta) \left( G_{\vartheta \vartheta} G_{\varphi \varphi} - G_{\vartheta \varphi}^2 \right)^{1/2},
\]

where the second derivatives have to be taken in the equilibrium position for which \( G_{\vartheta} = G_{\varphi} = 0 \). The expression does not always hold for \( \sin \vartheta = 0 \). We see that the resonance frequency is proportional to the geometrical mean stiffness in two perpendicular directions which coincide with the principal axes of the energy.

We shall now apply this equation (7) to the case of a uniaxial crystal of ellipsoidal form with the \( z \)-axis as the preferred direction. The demagnetization coefficients are \( N_x, N_y \) and \( N_z \). A static magnetic field \( H \) is applied
in the direction $\theta = \varphi = \pi/2$, and the sample is assumed to be homogeneously magnetized. The free enthalpy is then the sum of the crystalline anisotropy energy, the magnetostatic and the demagnetization energies, and reads

$$G = K \sin^2 \theta - HM \sin \theta \sin \varphi +$$

$$+ \left( M^3/2 \right) \left( N_x \sin^2 \varphi \cos^2 \theta + N_y \sin^2 \varphi \sin^2 \theta + N_z \cos^2 \theta \right). \quad (8)$$

The equilibrium orientation is given by

$$\sin \theta = \frac{H}{H^4 + (N_y - N_z)M}, \quad \varphi = \pi/2$$

for $$(N_y - N_x) M < H < H^4 + (N_y - N_z)M \quad (9)$$

and $\theta = \varphi = \pi/2$ for $H$ exceeding the latter value. Using (7) we then find for a spheroid with $N_x = N_y = N$ and $N_z = 4\pi - 2N$ for the resonance conditions

$$(\omega_r/\gamma)^2 = \begin{cases} H^4 - (4\pi - 3N)M^2 - H^2 & H < H^4 - (4\pi - 3N)M, \\ x & H > H^4 - (4\pi - 3N)M. \end{cases} \quad (10)$$

The first curve is a circle and the second one a hyperbola (see fig. 1, dashed curve).

Fig. 1. Dependence of the resonance frequency on the strength of the applied magnetic field $H$ perpendicular to the hexagonal axis for various orientations of the microwave field $h$ and of the Bloch walls with respect to the static field. The values of the angle $\alpha$ between the Bloch walls and $H$ are indicated ($0, \pi/2$). For $H > H^4 + NM$ the specimen is saturated in the $y$-direction. The $\omega_r/\gamma$- and $H$-scales are the same. The graph applies approximately to BaFe$_{12}$O$_{19}$ at room temperature.
3. Influence of Weiss-domain structure

If there is no static magnetic field present, the ellipsoid will be divided into Weiss domains, with the Bloch walls parallel to the preferred axis. This Weiss-domain structure will persist on the application of a perpendicular field, and will presumably not be changed very much. The field only rotates all spins in the domains. The resultant magnetization in the z-direction remains zero, so that there are no demagnetizing fields in this direction. Moreover the component of the magnetization perpendicular to the walls is continuous (no magnetic poles on the walls). It has been shown 2) that this does not necessarily remain the case for the dynamic component of the magnetization vector. In order to investigate this for the present case we write down the expression for the free enthalpy which is now a function of the orientations ($\theta_1$, $\varphi_1$) and ($\theta_2$, $\varphi_2$) of the two magnetization vectors of the two kinds of Weiss domains. This energy per cm$^3$ reads

$$G = \left(\frac{K}{2}\right) (\sin^2 \theta_1 + \sin^2 \theta_2) - \left(\frac{H M}{2}\right) (\sin \theta_1 \sin \varphi_1 + \sin \theta_2 \sin \varphi_2) +$$
$$+ \left(\frac{M}{2}\right) \left[(N/4) (\sin \theta_1 \cos \varphi_1 + \sin \theta_2 \cos \varphi_2)^2 +
+ (N/4) (\sin \theta_1 \sin \varphi_1 + \sin \theta_2 \sin \varphi_2)^2 + (\pi - N/2) (\cos \theta_1 + \cos \theta_2)^2 +\right.$$  
$$\left.\tau \sin \theta_1 \cos (\varphi_1 - \alpha) - \sin \theta_2 \cos (\varphi_2 - \alpha) \right].$$  

(11)

It is assumed that the Weiss domains are thin slabs, as is suggested by the powder pattern of fig. 2, where the surface is in a basal plane. It is taken on a much thicker crystal.

Fig. 2. Powder pattern of a BaFe$_{12}$O$_{19}$ single crystal. The surface is a basal plane. Magnification 350 ×.
The last term of (11) represents the demagnetization on the walls of these very thin Weiss domains (with demagnetization coefficient \(4\pi\)) and is equal to

\[2\pi \left( \frac{M_{1n} - M_{2n}}{2} \right)^2,\]

where \(M_{1n}\) and \(M_{2n}\) are the components of \(M_1\) and \(M_2\) normal to the wall. The angle between the Bloch wall and the static field is \(\alpha\). The equilibrium position is given by

\[
\sin \theta_1 = \sin \theta_2 = H/(H^A + NM), \quad \theta_1 + \theta_2 = \pi, \quad \varphi_1 = \varphi_2 = \pi/2, \quad H < H^A + NM. \tag{12}
\]

The value of \(\sin \theta\) corresponds to that of (9) with \(N_z = 0\). In this case the binding to the \(z\)-axis is always increased by the demagnetization.

Proceeding in the same way as in section 2 one now gets a four by four matrix for the equations of motion. The problem has some resemblance to that of antiferromagnetic resonance \(^3\). In our case the two magnetization vectors are coupled by demagnetizing fields, instead of by Weiss molecular fields.

For convenience we put \(\Delta \theta_1 = x_1, \Delta \varphi_1 = x_2, \Delta \theta_2 = x_3, \Delta \varphi_2 = x_4\) and \(\omega M \sin \theta/\gamma = z\), with \(\vartheta = \theta_1 = \pi - \theta_2\). In the experiments the a.c. magnetic field was always in the basal plane; so we confine ourselves to this case and we let \(\beta\) denote the angle between the a.c. field \(h\) and the static field.

The equations of motion are then

\[
\begin{align*}
G_{11}x_1 + (G_{12} + iz/2)x_2 + G_{13}x_3 + G_{14}x_4 &= -(M/2)h \cos \beta \cos \theta, \\
(G_{21} - iz/2)x_1 + G_{22}x_2 + G_{23}x_3 + G_{24}x_4 &= (M/2)h \sin \beta \sin \theta, \\
G_{31}x_1 + G_{32}x_2 + G_{33}x_3 + (G_{34} + iz/2)x_4 &= (M/2)h \cos \beta \cos \theta, \\
G_{41}x_1 + G_{42}x_2 + (G_{43} - iz/2)x_3 + G_{44}x_4 &= (M/2)h \sin \beta \sin \theta,
\end{align*}
\]

where \(G_{ij} = \partial^2 G/\partial x_i \partial x_j\). We have \(G_{11} = G_{33}, G_{22} = G_{44}\) and \(G_{12} = -G_{14} = G_{23} = -G_{24}\). It is convenient to use as variables \(\Delta \theta^\pm = (\Delta \theta_1 \pm \Delta \theta_2)/2\) and \(\Delta \varphi^\pm = (\Delta \varphi_1 \pm \Delta \varphi_2)/2\). One then gets

\[
\begin{align*}
A \Delta \theta^+ + iz \Delta \varphi^+ - n \Delta \varphi^- &= 0, \\
B \Delta \theta^- + iz \Delta \varphi^- &= -Mh \cos \beta \cos \theta, \\
C \Delta \varphi^+ - iz \Delta \theta^+ &= Mh \sin \beta \sin \theta, \\
D \Delta \varphi^- - iz \Delta \theta^- &= 0,
\end{align*}
\]

where

\[
\begin{align*}
A &= 2(G_{11} + G_{13}) = M[(H^A + 4\pi M \sin^2 \alpha) \cos^2 \vartheta + (4\pi - 2N) M \sin^2 \vartheta], \\
B &= 2(G_{11} - G_{13}) = M (H^A + NM) \cos^2 \vartheta, \\
C &= 2(G_{22} + G_{24}) = M (H^A + NM) \sin^2 \vartheta, \\
D &= 2(G_{22} - G_{24}) = M (H^A + 4\pi M \cos^2 \vartheta) \sin^2 \vartheta, \tag{14}
\end{align*}
\]
whereas the non-diagonal element \( n \) is given by
\[
n = -4G_{12} = 2\pi M^2 \sin \theta \cos \theta \sin 2\alpha.
\] (15)

For \( n = 0 \), which is the case for walls parallel or perpendicular to the static field, (13) splits into two separate sets of equations formed by the elements of the first and third rows and by those of the second and the fourth ones respectively, which give resonance frequencies obeying \( z_r^2 = AC \) and \( z_t^2 = BD \). It is seen that the first resonance mode can only be excited for \( \sin \beta \neq 0 \), i.e. only by the component of the a.c. magnetic field which is normal to the static magnetic field. The second resonance mode can only be excited by the parallel component. It can be seen that an a.c. field in the \( z \)-direction excites the same resonance mode as that in the \( x \)-direction. In that case the right-hand side of the first equation of (13) contains \( h_z \).

In the more general case of \( n \neq 0 \) the two resonance modes are mixed up. The resonance frequencies then obey
\[
z_r^2 = \frac{AC + BD}{2} \pm \sqrt{\left(\frac{AC - BD}{2}\right)^2 + n^2BC^2}.
\]

For \( \text{BaFe}_{12}\text{O}_{19} \), \( n \) is comparatively small at the frequency used, so that
\[
\left(\frac{\omega}{\gamma}\right)_1 \approx (H^A + NM) (H^A + 4\pi M \sin^2 \alpha) + \frac{H^A + 4\pi M \sin^2 \alpha - (4\pi - 2N)M}{H^A + NM} H^2 + \Delta,
\] (16a)
\[
\left(\frac{\omega}{\gamma}\right)_2 \approx (H^A + NM) (H^A + 4\pi M \cos^2 \alpha) + \frac{H^A + 4\pi M \cos^2 \alpha}{H^A + NM} H^2 - \Delta,
\] (16b)

with
\[
\Delta = \frac{n^2BC}{(AC - BD)M^2} \sin^2 \theta.
\] (17)

In (16) also we have labelled \( (\omega/\gamma)^2 \) with the subscripts 1 and //, since the amount of mixing is small. The intensity of the absorption is proportional to the susceptibility for different excitations of the same mode. The a.c. component of the magnetization in the direction of \( h \) is equal to \( M \left\{ A \delta^- \cos \theta \cos \beta - \Delta \varphi^+ \sin \theta \sin \beta \right\} \) and can be calculated from (13). One then finds for the susceptibility a fraction whose numerator for e.g. \( \beta = \pi/2 \) is proportional to \( \sin^2 \theta \left\{ A (BD - z^2) - n^2B^2 \right\} \). This indeed does not vanish at the parallel resonance frequency, but is equal to \( n^2B^2 \sin^2 \theta/(AC - BD) \).

We have to compare this with the value for \( \beta = 0 \), i.e. \( \cos^2 \theta \left\{ D(AC - z^2) - n^2C^2 \right\} \) which is approximately \( \cos^2 \theta D(AC - BD) \). The ratio of the two susceptibilities is most 0.04 in the cases investigated for \( \alpha = \pm \pi/4 \).

For \( H > H^A + NM \) all magnetization vectors have arrived in the
direction of $H$, and the Weiss-domain structure has disappeared. We must then use the second resonance condition of (10):

\[(\omega_r / \gamma)^2 = H^d - H - (4\pi - 3N)M^d \quad H > H^d + NM. \quad (18)\]

For \( H = H^d + NM \) the two frequencies of (16a) and (18) for the transverse field are the same and satisfy

\[(\omega_r / \gamma)^2 = (H^d + NM)(4\pi - 2N)M \quad H = H^d + NM. \quad (19)\]

Only for the longitudinal microwave field does the resonance frequency become zero as in the simple case without Weiss-domain structure (fig. 1). The absorption peak then vanishes also, since it is generated by the component of the field perpendicular to the magnetization. For transverse excitation the resonance frequency does not vanish owing to the fact that for this mode $\Delta \theta_1$ and $\Delta \theta_2$ have the same sign so that the resultant a.c. magnetization is finite in the $z$-direction. Demagnetizing fields are then set up, resulting in the term $(4\pi - 2N)M$ in (16a) and (19), so that $\omega_r$ remains finite. It is impossible that a movement of the Bloch walls compensates for this a.c. magnetization, since the results of Wijn's experiments show that these walls are immobile at the high frequency used (24 000 Mc/s).

Fig. 1 shows the various curves obtained for $a = 0$ and $a = \pi/2$. It is seen that at a low, constant frequency, and varying $H$, only one absorption peak for the parallel a.c. field is obtained. If the frequency is increased to that of (19) one should expect both for the parallel and the transverse a.c. fields one absorption peak. For still higher frequencies the peak for the transverse a.c. field splits up into two peaks. All peaks for fields lower than $H^d + NM$ are broadened by the Weiss-domain structure.

It is also possible to obtain these different types of spectra at a constant frequency by varying the temperature, since the resonance frequency of (19) for $N = 0$ is proportional to $\gamma B K$ and $K$ decreases appreciably with increasing temperature for BaFe$_{12}$O$_{19}$ \(^1\). It appeared that at room temperature at 24 000 Mc/s we just have resonance in the minimum of the curve of fig. 1 for transverse excitation. Accordingly one should expect this second peak to disappear at low temperatures and to split into two peaks at elevated temperatures. This has been actually observed as will be reported in section 5.

4. Measuring technique

In order to prevent dimensional effects, the magnetic crystal should be small compared to the internal wavelength. Therefore a thin sheet of BaFe$_{12}$O$_{19}$ with lateral dimensions of about 1 mm was glued to a reflecting piston and the specimen was ground to 40 $\mu$ thickness. X-ray diffraction
showed that it was a proper single crystal. The hexagonal axis was normal to the plate. A piece of a $\frac{1}{2}$" by $\frac{1}{4}$" wave guide between the piston and a wall with a rectangular hole, i.e. an inductive iris, formed a resonant cavity which can be tuned to 24 000 Mc/s by the adjustment screw of the piston (see fig. 3). The cavity can be exposed to a static magnetic field, which can be rotated in a plane normal to the crystal axis.

The cavity is coupled to a wave-guide system for the 1.25-cm region, so that it resembles a measurement equipment for paramagnetic resonance, frequently described in present-day literature \(^5\). By the application of a directional coupler C, equilibrium can be established with zero output in the heterodyne detector if the resonant cavity is adapted to the guide. This can be carried out by tuning the cavity to resonance with piston $P_1$ and adjusting the proper coupling with $P_2$. Small resonance absorption in the crystal gives a small decrease in $Q$, giving a reflected wave resulting in a voltage $V$ proportional to the absorption i.e. to the imaginary part of the magnetic moment. This signal is measured with a superheterodyne device, consisting of a local oscillator klystron $D_L$, mixing crystal $D_M$, intermediate-frequency amplifier $D_B$, rectifier, and d.c. meter. Another necessary component of the system is the frequency stabilization described by Pound \(^6\). This makes it easy to eliminate dispersion, because the cavity
can be tuned properly for each measurement by adjusting the frequency till minimum output is reached.

The temperature of the cavity can be raised up to 200 °C by a heating filament and is measured by a thermocouple. For measurements at the temperature of liquid nitrogen the cavity is put into a Dewar flask. In this case a big Oerlikon electromagnet was used with an air gap of 70 mm. In other cases an air gap of 14 mm in a Boas magnet, producing a maximum field of 20 000 Oe was sufficient. The magnetic field was calibrated by proton resonance and it appeared that hysteresis had to be taken into account.

5. Experimental results

Absorption measurements were made at five different temperatures as a function of the magnitude of the magnetic field. The direction of the field makes an angle of 45° with the h.f. magnetic field, the latter being in the direction of the longer dimension of the wave guide and tangential to the reflecting surface of the piston. In this position the components of the a.c. field parallel and perpendicular to the d.c. field are of equal magnitude. Figs 4a, b, c, d, and e show the absorption of the crystal specimen at about 24 000 Mc/s, for the same input energy level, for temperatures —196, 20, 112, 155 and 200 °C. The ordinate represents the voltage \( V \) induced by the reflected wave, in arbitrary units, but the scale is the same at all temperatures. The low-field peak is only excited by a parallel a.c. field and the high-field peaks by a transverse a.c. field, as is shown in fig. 5 for room temperature and as was established also at the other temperatures. This confirms the expectation of the-theory of section 3. At low temperatures only the parallel-field peak is found, but at higher temperatures the perpendicular-field peak comes up and splits into two at the highest temperature. The mean values of \( H \) of the absorption peaks are listed in table I, together with the values of the measuring frequency, of \( 4\pi M \) and \( NM \).

### Table I

<table>
<thead>
<tr>
<th>( T ) (°C)</th>
<th>( H_{\text{res}} ) (kOe)</th>
<th>( f ) (Mc/s)</th>
<th>( 4\pi M ) (kgauss)</th>
<th>( NM ) (kOe)</th>
</tr>
</thead>
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<tr>
<td>-196</td>
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<td>23 930</td>
<td>6·67</td>
<td>0·20</td>
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<td>0·14</td>
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<td>3·50</td>
<td>0·10</td>
</tr>
<tr>
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<td>15·35</td>
<td>18·15</td>
<td>3·12</td>
<td>0·09</td>
</tr>
</tbody>
</table>
Fig. 4. Absorption peaks as a function of the strength of the static field $H$ at five different temperatures on the same (arbitrary) scale with the microwave field in the basal plane at $45^\circ$ with the static field $H$. 
6. Evaluation of \( g \) and \( H^A \)

\( T = 20 ^\circ C \). If we start with the room-temperature measurements, we assume that there we are just in the minimum of the curve for transverse excitation of fig. 1. This point is not sharp, since the sample is not an ellipsoid but a disc, so that the demagnetization is not homogeneous. Thus \( H^A + NM = 17.15 \text{ kOe at room temperature or } H^A = 17.0 \text{ kOe.} \) We now apply equation (16b) with \( \alpha = \pi/4 \) as a mean value for the low-field absorption peak. One then finds \( \omega_r/\gamma = 8.65 \text{ kOe or } g = 1.98. \) The minimum of \( \omega_r/\gamma \) for transverse excitation is, according to (19), equal to 8.78 kOe, so that the resonance absorption we actually find for transverse excitation exists only by virtue of the inhomogeneity of the demagnetization. The peak height is therefore small.

\begin{align*}
T = 20 ^\circ C
\end{align*}

[Image: Absorption peaks at 20 ^\circ C as a function of \( H \) both with the microwave magnetic field parallel and perpendicular to the static field \( H \).

\( T = 200 ^\circ C \). At this temperature the three peaks are well resolved. Subtraction of equation (16b) from (16a) gives \( H^A + NM \) in terms of the resonance fields \( H_{//} \) and \( H_\perp \) for \( \alpha = \pi/4\):

\begin{equation}
H^A + NM = \frac{(2\pi - N) M(H_{//}^2 + H_\perp^2)}{H_{//}^2 - H_\perp^2 - \Delta_{//} - \Delta_\perp}.
\end{equation}

\( \Delta_\perp \) and \( \Delta_{//} \) are small quantities which can be estimated at once with sufficient accuracy. One then finds \( H^A + NM = 16.2 \text{ kOe, which is smaller than } H_\perp \text{ (16.85 kOe). Obviously } (H_\perp - H_{//}) \text{ is too large. A possible explanation of this discrepancy might be a disorientation of the crystal with respect to the static magnetic field. The effect thereof has been investigated in the Appendix. The result is that the two resonance peaks}
below saturation are not seriously affected and at most approach each other slightly, this being contrary to the experimental finding. Only the high-field peak is shifted towards a lower value of the field. We shall assume therefore that the effect of disorientation on the resonance conditions is negligible, as we should expect from the accuracy of mounting.

The resonance peaks found are very broad, much broader than would be expected from the Weiss-domain structure. It is possible therefore that the two transverse resonance peaks are not well resolved, and in any case have decreased their distance apart. This can apply quite well to the spectra at 112 and 155 °C, but we shall assume that also at 200 °C the two transverse resonance peaks are shifted over distances in the $H$ scale which are proportional to their difference with $(H^d + NM)$ and of opposite sign. Other plausible assumptions do not give, however, substantially different results. We then obtain for $H^d + NM = 17·40$ kOe, i.e., $H^d = 17·3$ kOe, $g = 2·02$ and $H_\perp = 16·70$ and 18·40 kOe, as compared with the experimental values of 16·85 and 18·15 kOe.

$T = 155$ °C. Also in this case (20) gives too low a value for $H^d + NM$ (15·8 kOe). For $H^d + NM = 17·45$ kOe or $H^d = 17·3$ kOe one finds from the parallel excitation $g = 2·01$. The values of the fields for transverse resonance are then 16·90 and 18·25 kOe, as compared with the observed values of 17·15 and 17·90 kOe. The differences between the calculated and the experimental values of the resonance fields are here larger than at $T = 200$ °C, owing to the greater overlap of the peaks.

$T = 112$ °C. The best fit with the experimental values can be obtained by taking $H^d + NM = 17·45$ kOe or $H^d = 17·3$ kOe. From the value of $H_{\text{res}}$ for the parallel excitation one then obtains $\omega r/\gamma = 8·57$ kOe or $g = 2·00$. For the transverse excitation the resonance fields are then found to be 17·15 and 17·95 kOe.

$T = -196$ °C. Since one cannot deduce both $H^d$ and $g$ from a single resonance peak, we assume $g$ to be 2·00, and find then $H^d = 16·2$ kOe.

### TABLE II

Calculated values of the anisotropy field $H^d = 2 K/M$ and the spectroscopic splitting factor $g$

<table>
<thead>
<tr>
<th>$T$ (°C)</th>
<th>$H^d$ (kOe)</th>
<th>$g$</th>
</tr>
</thead>
<tbody>
<tr>
<td>-196</td>
<td>16·2</td>
<td>2·00 (assumed)</td>
</tr>
<tr>
<td>20</td>
<td>17·0</td>
<td>1·98</td>
</tr>
<tr>
<td>112</td>
<td>17·3</td>
<td>2·00</td>
</tr>
<tr>
<td>155</td>
<td>17·3</td>
<td>2·01</td>
</tr>
<tr>
<td>200</td>
<td>17·3</td>
<td>2·02</td>
</tr>
</tbody>
</table>
The minimum value of $\omega_r/\gamma$ for transverse excitation is, according to (19), equal to 10.1 kOe, which is well above that of the measuring frequency, so that the second peak cannot occur.

The results obtained are collected in table II.

We do not consider the differences found for $g$ at different temperatures to be significant; they are within the experimental error. We conclude therefore that the $g$-factor of BaFe$_{12}$O$_{19}$ has the spin-only value of 2.00 with an accuracy of $\pm 1\%$ at all temperatures.

7. Shape and intensities of the absorption lines

In the first place we see that the parallel resonance peaks are asymmetrical in the sense that their maximum is shifted to lower fields. This can be explained by the occurrence of the term $A$ in (16b) which is maximum for $\alpha = \pi/4$ and zero for $\alpha = 0$ or $\pi/2$. For the transverse peak, $A$ is negligible. Secondly the width of the parallel-field resonance peaks decreases with increasing temperature. This fact is understandable if the width is caused by the Weiss-domain structure in which case it should be proportional to $M$.

We shall now calculate the areas of the absorption peaks. For this we shall assume that the peaks are caused by separate single resonance phenomena occurring at $\omega_r(H)$. Consequently the non-diagonal elements $n$ in (13) and with them $A$ in (16a) and (16b) will be ignored.

From one of the Kronig-Kramers relations between the real and the imaginary parts of the susceptibility, i.e.

$$\chi'(\omega) = \frac{1}{\pi} \int_0^\infty \frac{\omega_1 \chi''(\omega_1)}{\omega_1^2 - \omega^2} \, d\omega_1,$$

follows, if it is assumed that the integrand is only finite for values of $\omega_1$ which are slightly different from that of $\omega_r$:

$$\pi(\omega_r^2 - \omega^2) \chi'(\omega) = \frac{d\omega_r^2}{dH} \int_0^\infty \chi''(H) \, dH. \quad (21)$$

If the damping is small, as has been assumed, $\chi'$ is substantially independent of it, and can be calculated at once from (5) by adding to the right-hand sides of these equations terms due to the microwave field, as in (13). If $\chi$ refers to the $\xi$-direction, one finds

$$\chi'_{\xi\xi}(\omega) = \frac{\gamma^2 G_{\eta\eta}}{(\omega_r^2 - \omega^2)}, \quad (22)$$

so that (21) becomes

$$\int_0^\infty \chi''_{\xi\xi}(H) \, dH = \frac{\pi G_{\eta\eta}}{d(\omega_r/\gamma)^2/dH^{-1}}. \quad (23)$$
For the case that \( G \) is only due to a static field \( H \), (23) reduces to \( \pi M/2 \).

For the parallel excitation, \( G_{\parallel} \) is, according to (13) and (14), given by
\[
D/\sin^2\vartheta, \text{ which is, for } \alpha = \pi/4, \text{ equal to } M\left[H^4 + 2\pi M\right].
\]
The derivative of \((\alpha r/\gamma)^2\) with respect to \( H \) is easily calculated from (16b) so that one gets, after multiplication by \( \cos^2\vartheta \) in order to take into account that only the component of the microwave field normal to the magnetization is active and that only the component of the a.c. magnetization parallel to the a.c. field is detected:
\[
\int_0^\infty \chi''_{\parallel}(H) dH = \frac{\pi M}{2} \left\{ \frac{H^4 + NM}{H_{\parallel}} - \frac{H_{\parallel}}{H^4 + NM} \right\},
\]
where \( H_{\parallel} \) denotes the field strength for parallel-field resonance.

For the perpendicular excitation, \( G_{\perp} \) is given by \( A \) of (13) and (14) so that we get for the first transverse resonance peak
\[
\int_0^{H^4 + NM} \chi''_{\perp}(H) dH = \frac{\pi M}{2} \left\{ \frac{(H^4 + NM)^2}{H_{\perp}(H^4 - 2\pi M + NM)} - \frac{H_{\perp}}{H^4 + 2\pi M} \right\},
\]
whereas for the second one we get
\[
\int_{H^4 + NM}^\infty \chi''_{\perp}(H) dH = \frac{\pi M}{2} \left[ \frac{H_{\perp} - H^4 + (4\pi - 3N)M}{2H_{\perp} - H^4 + (4\pi - 3N)M} \right].
\]
We have calculated the factor of \( \pi M/2 \) for the various resonance peaks (table III). The field strengths used in (25) and (26) are the calculated ones.

### TABLE III

Calculated line intensities \( I \) in units \( \pi M/2 \)

<table>
<thead>
<tr>
<th>( T (\circ C) )</th>
<th>( I_{\parallel} )</th>
<th>( I_{\perp} )</th>
</tr>
</thead>
<tbody>
<tr>
<td>-196</td>
<td>0.27</td>
<td></td>
</tr>
<tr>
<td>20</td>
<td>0.26</td>
<td></td>
</tr>
<tr>
<td>112</td>
<td>0.25</td>
<td>0.25</td>
</tr>
<tr>
<td>155</td>
<td>0.25</td>
<td>0.26</td>
</tr>
<tr>
<td>200</td>
<td>0.25</td>
<td>0.26</td>
</tr>
</tbody>
</table>

The peak height for parallel excitation is practically constant. According to table III the intensity is proportional to \( M \), and this is consistent with
a constant peak height, as follows from the discussion in the beginning of this section. The area of the peak at the lowest transverse field should, according to table III, be equal to that of the parallel-field peak. Experimentally it is somewhat larger; since the width is comparable, but the height is greater. This is presumably connected with the shift of the resonance lines, which was discussed in the preceding section. The highest-field peak should, according to table III, have a larger area. The height is the same, but at 200 °C the width is certainly greater. The general agreement is therefore satisfactory.

8. Discussion

The above analysis was based upon the validity of the use of equation (2), i.e. no terms with higher power than the second of \( \sin \theta \) have been considered. This has been established experimentally by Mr P. Jongenburger of this laboratory, at temperatures below room temperature, with an accuracy of about 1% by measuring the magnetization curve in the difficult direction. This is contrary to what is found for other hexagonal crystals \(^8\) such as Co, MnBi and MnSb, where the second anisotropy constant is appreciable and at least of the order of twenty or forty per cent of the first anisotropy constant.

We have seen that the g-factor of BaFe\(_{12}\)O\(_{19}\) has the spin-only value, so that there is no contribution of orbital motion to the magnetic moment. Roughly this is what one would expect, since the magnetic moments are those of the ferric ions, which have a \( ^6S \) ground state. In this respect one can compare BaFe\(_{12}\)O\(_{19}\) with MnFe\(_2\)O\(_4\), for which substance \( g \) is also equal to 2.00. We are therefore forced to believe that spin-orbit interaction is inactive in BaFe\(_{12}\)O\(_{19}\). Mostly, however, the occurrence of crystalline anisotropy is ascribed to spin-orbit interaction, and this anisotropy is appreciable here. Another cause for crystalline anisotropy in uniaxial crystals is dipole-dipole interaction, but then one can expect a term with \( \sin^2 \theta \) only, the other ones being exactly zero. Since this agrees with experimental finding, we regard it as highly probable that the magnetism in BaFe\(_{12}\)O\(_{19}\) originates from electron spins only, and that the anisotropy is caused by dipole-dipole interaction. Preliminary calculations however, do not yet support the latter point of view.

We wish to thank Mr L. van der Kint for his assistance during the measurements.

Eindhoven, February 1955
Appendix

For the case that there is a component of the field $H_1$ along the hexagonal axis, we assume that the domain boundaries shift in such a way that the mean static magnetic field inside the specimen along the easy direction remains zero, i.e.

$$H_1 - (4\pi - 2N)\left\{(1 + v) \cos \vartheta_1 - (1 - v) \cos \vartheta_2\right\} M/2 = 0, \quad (A1)$$

where $(1 \pm v)/2$ are the total volumes of the two kinds of Weiss domains. The equilibrium orientation of the magnetic vectors is therefore unchanged and is given again by (12) so that (A1) yields

$$v = H_1/(4\pi - 2N)M \cos \vartheta. \quad (A2)$$

The free enthalpy now becomes instead of (11):

$$G = (K/2) \left\{ \sin^2 \vartheta_1 (1 + v) + \sin^2 \vartheta_2 (1 - v) \right\} +
- (HM/2) \left\{ \sin \vartheta_1 \sin \varphi_1 (1 + v) + \sin \vartheta_2 \sin \varphi_2 (1 - v) \right\} +
- (H_1M/2) \left\{ \cos \vartheta_1 (1 + v) + \cos \vartheta_2 (1 - v) \right\} +
+ \left( M^2/2 \right) \left\{ (N/4) \sin \vartheta_1 \cos \varphi_1 (1 + v) + \sin \vartheta_2 \cos \varphi_2 (1 - v) \right\}^2 +
+ \left( \pi - N/2 \right) \left\{ \cos \vartheta_1 (1 + v) + \cos \vartheta_2 (1 - v) \right\}^2 +
\left( \pi \right) \sin \vartheta_1 \cos (\varphi_1 - \alpha) - \sin \vartheta_2 \cos (\varphi_2 - \alpha) \right\}^2 (1 - v^2)] . \quad (A3)$$

The equations of motion, expressed in the variables $\frac{1}{2} \{ x_1 (1 + v) \pm x_3 (1 - v) \}$ and $\frac{1}{2} \{ x_2 (1 + v) \pm x_4 (1 - v) \}$ yield the following determinantal equation for $\omega_r$:

$$| A - g_{12} v \quad iZ_r + nv \quad -n |
| g_{21} v \quad B \quad 0 \quad iZ_r |
| -iZ_r \quad 0 \quad C \quad 0 |
| -n \quad -iZ_r + nv \quad g_{43} v \quad D | = 0$$

in which

$$g_{12} = (M \cos \vartheta)^2 \left( -4\pi \sin^2 \alpha + N \right) ,$$
$$g_{21} = (M \sin \vartheta)^2 \left( 4\pi - 2N \right) ,$$
$$g_{43} = (M \sin \vartheta)^2 \left( -4\pi \cos^2 \vartheta + N \right) .$$

The situation becomes most simple for $N = 0$, which is approximately satisfied for our sample $(N/4\pi = 0.03)$. One then obtains

$$(\omega_r/\gamma)^2 = H^4 \left[ H^4 \cos^2 \vartheta + 2\pi M \right] \left\{ 1 \pm \sqrt{\cos^2 2\vartheta + \sin^2 2\vartheta \sin^2 \alpha (1 - v^2)} \right\}.$$
curves for $a = 0$. They meet them at the same value of $H$, corresponding to $v = 1$, both for longitudinal and for transverse excitation. The resonance frequency for parallel field is there still finite.

Then, again, the transverse curve continues with a different slope. This curve can be calculated for the case where the field $H$ makes an angle $(\pi/2 - \varepsilon)$ with the hexagonal axis in which $\varepsilon \ll 1$. It is then found that $(\omega_r/\gamma)^2$ is increased with respect to (18) by an amount of approximately

$$A(\omega_r/\gamma)^2 = \varepsilon^2 H_i H^2 - (4\pi - 3N) M_i^2 (5h_r^2 - 6h_r + 2)/2(h_r - 1)^2,$$

in which

$$h_r = H/H^2 - (4\pi - 3N) M_i^2.$$