Antiferromagnetism and Antiferromagnetic Resonance in a Rhombic Crystal at $T=0$

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Neél's phenomenological theory of antiferromagnetism at zero temperature has been worked out for a three-dimensional case. When the external magnetic field comes near to a certain hyperbola in the field space lying in the plane through the favored and the least favored direction of magnetization, an anomaly occurs. This anomaly consists of a rapid turn-over of the two opposed directions of magnetization into the direction perpendicular to the plane just mentioned. At much higher fields the two opposed directions of magnetization will deviate towards the field, and finally antiferromagnetism will go over into paramagnetic saturation.

Following the general method of Kittel, Ubbink has investigated antiferromagnetic resonance. At not too high frequencies two resonance fields will be flanking the hyperbola in field space. At higher frequencies one resonance field will decrease and vanish, while the other one will approach the field of paramagnetic resonance.

IN Neél's phenomenological description of antiferromagnetism[1] the magnetic ions are divided over two sublattices, which we shall indicate, respectively, by a single and by a double dash.

In addition to the extermal field H, the spins in the first sublattice experience a virtual magnetic field $-\alpha\sigma''$, while the spins in the double dashed sublattice experience a virtual field $-\alpha\sigma'$, where the σ's represent the average magnetization in the sublattice indicated.

Now the virtual fields tend to keep the magnetizations of the two sublattices exactly opposite to each other. As long as the external field is small compared to the virtual fields it tends to orientate the opposite magnetizations perpendicularly to itself, since then the magnetic decrease of energy is largest. On the other hand, crystalline anisotropy may favor certain orientations of the opposite magnetizations. When the external field becomes of the same order as the virtual fields, it forces the magnetizations into its own direction. In the investigation discussed the interplay of these different tendencies is analyzed.

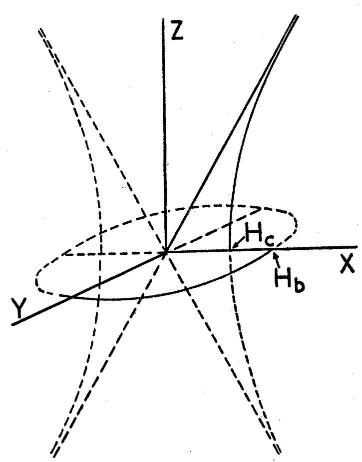

FIG. 1. Hyperbola and ellipse in the H space.

Let us suppose that α as well as μ differ in the direction of the three rhombic axes x, y, and z. Considering ionic levels with a twofold degeneration we get

$$H_x'=H_x-(\alpha_x\mu_x H_x''/W'')\tanh(W''/kT)(+\text{perm}),$$
$$H_x''=H_x-(\alpha_x\mu_x H_x'/W')\tanh(W'/kT)(+\text{perm}),$$

where

$$W'=+(\mu_x^2H_x'^2+\mu_y^2H_y'^2+\mu_z^2H_z'^2)^{\frac{1}{2}},$$
$$W''=+(\mu_x^2H_x''^2+\mu_y^2H_y''^2+\mu_z^2H_z''^2)^{\frac{1}{2}}.$$

In cooperation with Dr. J. Haantjes, professor of geometry in Leiden, the solutions of these eight equations have been analyzed for $T=0$, in which case the hyperbolic tangents are equal to one.[2]

Four different solutions are found which will be indicated by A, B, C, and D. At zero external field the A, B, and C solutions give saturated sublattices magnetized in the $\pm x$, $\pm y$, and $\pm z$ directions, respectively. The D solution has an identical magnetization of the sublattices, and this represents the case of paramagnetic saturation. Up to an external magnetic field of the order of $\alpha\mu$ only the solutions A, B, and C exist. If $\alpha_x\mu_x^2>\alpha_y\mu_y^2>\alpha_z\mu_z^2$, which means that the $\pm x$ directions are energetically the most favorable ones and the $\pm z$ directions the least favorable ones, the A solution has and keeps the lower energy and will thus be realized. Between a field of the order of $\alpha\mu$ and one of the order

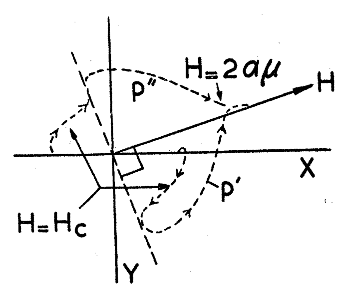

FIG. 2. The directions p' and p'' of the magnetizations of the sublattices when a field H, orientated in the xy plane, is increased.

[1] L. Neél, Ann. phys. 18, 5 (1932); 5, 232 (1936); 3, 137 (1948). J. H. Van Vleck, J. Chem. Phys. 9, 85 (1941).

[2] C. J. Gorter and J. Haantjes, Leiden Comm. Suppl. 104b, Physica 18, 285 (1952).

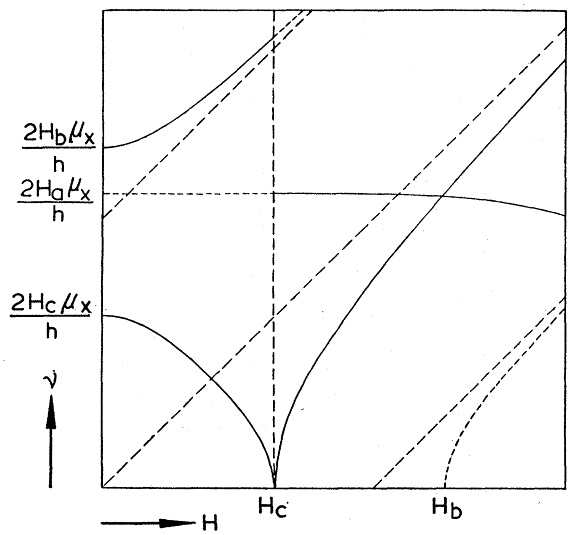

FIG. 3. Antiferromagnetic resonance frequencies drawn as a function of a field H in the x direction of the crystal. H_c is the turn-over field, and the dotted line through (0,0) gives the frequencies of paramagnetic resonance.

of $2\alpha\mu$ the magnetizations are strongly bent towards the field. In this region A, B, and D exist, while A remains the stable solution. Finally, the directions of the magnetizations in the three solutions merge, and the D solution is the only one that remains.

However, in the low field region certain very remarkable anomalies are to be expected. These anomalies occur when the field comes near to a certain hyperbola in the xz plane or to a certain ellipse in the xy plane. See Fig. 1. The anomaly then consists in a rapid exchange of place between the A and B solutions and the B and C solutions, respectively. In the neighborhood of the hyperbola we thus have, in the realized A solution, a rapid turn-over from directions near to $\pm x$ to directions near to $\pm y$. In this way the crystalline preference for magnetization in the $\pm x$ directions is checked by the magnetic field favoring directions perpendicular to itself. The turn-over field in the x direction is

$$H_c=(\alpha_x^2\mu_x^4-\alpha_y^2\mu_y^4)^{\frac{1}{2}}/\mu_x.$$

If the anisotropies are relatively small, we may say that the turn-over field

$$H_c=[2\alpha(\alpha_x\mu_x^2-\alpha_y\mu_y^2)]^{\frac{1}{2}}.$$

If the field lies in the xy plane, the turn-over takes place gradually in the same field region. If it lies in the xz plane, the magnetizations move gradually towards the $\pm c$ direction; but if the angle between field and x axis is smaller than $\arctan[\mu_x^2(\alpha_y^2\mu_y^4-\alpha_z^2\mu_z^4)/\mu^2(\alpha_x^2\mu_x^4-\alpha_y^2\mu_y^4)]^{\frac{1}{2}}$, a turn-over into the b direction occurs when the hyperbola in the field space is reached.

Figure 2 indicates how, as a result of all this, the two directions of magnetization move when a field, orientated in the xy plane, is gradually increased.

Following the general method of Bloch and Kittel,[3] Mr. J. Ubbink has extended the treatment given above to the antiferromagnetic resonance occurring when an alternating field is added.[4] If an external field is orientated in the x direction, resonance is, in general, to be expected at two frequencies (Fig. 3). In a very small field one of these is equal to $2H_c\mu_x/h$ and the other one is equal to $2H_b\mu_x/h$, where H_b is the field where the x axis cuts the ellipse in the xy plane. The lower one of these frequencies decreases with increasing field and becomes zero at $H=H_c$.

The larger frequency increases at the same time. At $H=H_c$ the turn-over takes place and the resonance frequencies are replaced by two others, one of which starts at zero and increases rapidly with increasing field, approaching the frequency of paramagnetic resonance $2H\mu_x/h$.

In practice the resonances are studied at constant frequency and varying field. As long as the frequency is relatively low we expect one resonance field slightly below and one slightly above H_c. The interval between the two will increase with increasing ν, and the low resonance field will become zero at $\nu=2H_c\mu_x/h$.

If H is having another position in the xz plane the hyperbola in field space will be flanked by two resonance curves, while in the xy plane the two resonances will merge and disappear when the angle with the x direction increases.

We shall not discuss now Ubbink's conclusions on the resonances in the higher fields of the order of $2\alpha\mu$.

Part of the conclusions of the calculations made in Leiden are identical with those obtained by Neél, Kittel, Nagamiya,[5] and recently by Yosida.[6]

[3] F. Bloch, Phys. Rev. **70**, 460 (1946); C. Kittel, Phys. Rev. **71**, 270 (1947); F. Keffer and C. Kittel, Phys. Rev. **85**, 329 (1952).
[4] J. Ubbink, Phys. Rev. **86**, 567 (1952).
[5] T. Nagamiya, Prog. Theoret. Phys. **6**, 350 (1951).
[6] K. Yosida, Prog. Theoret. Phys. **7**, 25 (1952).