## NATURE OF LOW-TEMPERATURE MAGNETIC ANOMALIES IN FERRITES WITH COMPENSATION POINTS

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By consideration of the nature of the temperature dependence of the spontaneous magnetization in gadolinium ferrite and in lithium ferrite-chromite, one can arrive at a condition for maximum probability of the appearance in ferrites of a point of compensation of the magnetic moments of the sublattices, or of an anomalous temperature variation of the spontaneous magnetization in the low-temperature region. The condition is this: one of the magnetic sublattices must possess "strong" exchange interaction, and consequently a Weiss temperature variation of spontaneous magnetization, and a second must possess a "weak" exchange interaction, which leads (in the presence of antiferromagnetic coupling to the "strong" sublattice) to a smeared out temperature variation of the spontaneous magnetization. It is concluded that in ferrites with compensation points, one should observe low-temperature magnetic anomalies, corresponding to an abrupt change of the long-range magnetic order in the sublattice with the "weak" exchange interaction. This conclusion is confirmed by the experimental data (cf. <sup>[2]</sup> and <sup>[4]</sup>).

1. In studies of ferrite-garnets of rare-earth elements [1,2] and of lithium ferrite chromite [3,4] in the low-temperature region (liquid nitrogen and liquid helium), anomalies have been observed in the temperature variation of magnetic and nonmagnetic properties (magnetic susceptibility, magneto-striction, coercive force, electrical resistivity, etc.).

The nature of these anomalies has hitherto not been explained. In this article we wish to show that the occurrence of the anomalies mentioned is connected with the existence of "low-temperature" points, corresponding to a sudden change of the long-range magnetic order in a sublattice with weak exchange interaction. Such a point should be especially well exhibited in ferrites with a compensation temperature, since in them there is a sharp difference in the nature of the exchange interactions in the sublattices; this in fact is the reason for the occurrence in the ferrite of a point of compensation of the magnetic moments of the sublattices.

2. We will analyze the condition for occurrence of a compensation point, for example, in the ferrite  $3Gd_2O_3 \cdot 5Fe_2O_3$ .

This ferrite is known to have three sublattices. On one of these (sublattice c) are located the  $Gd^{3+}$  ions, on the other two (a and d) the  $Fe^{3+}$  ions. The last two sublattices, experiment shows,

may be regarded as a single sublattice, thanks to the strong exchange interaction between the  $Fe^{3+}$ ions. We shall therefore refer hereafter to gadolinium ferrite-garnet as a ferrite consisting of two sublattices, 'gadolinium'' and ''iron''. We consider what form the  $\sigma_{\rm S}(T)$  curves will have for each of these sublattices.

In order to find the form of the  $\sigma_{s}(T)$  curve for the iron sublattice, it is necessary to "switch off" the influence of the gadolinium sublattice in the magnetic sense, i.e., to replace all the  $Gd^{3+}$ ions with nonmagnetic ions, for example  $Y^{3+}$  or  $Lu^{3+}$  ions. Such a "substituted" ferrite is  $3Y_2O_3$ •5Fe<sub>2</sub>O<sub>3</sub> or 3Lu<sub>2</sub>O<sub>3</sub>•5Fe<sub>2</sub>O<sub>3</sub>. These ferrites, experiment shows, have a normal Weiss temperature variation of the spontaneous magnetization (Fig. 1, curve 1). Complete destruction of the magnetic long-range order occurs in them at a high temperature ( $\sim 560^{\circ}$  K), and this indicates the presence in the iron sublattice of strong exchange interaction. Knowing the approximate form of the temperature dependence of the spontaneous magnetization of the iron sublattice, we can determine, from the experimental  $\sigma_{s}(T)$  curve for gadolinium ferrite (Fig. 1, curve 3), the approximate form of the curve for the gadolinium sublattice (Fig. 1, curve 2). It is evident that the gadolinium sublattice has an extremely spread-out temperature variation of the spontaneous magnetization, for which it is difficult



even to define the position of the Curie point. Several alloys (for example 30% Cu, 70% Ni and others) have a similar type of  $\sigma_{\rm S}(T)$  curve.

One asks, why is the gadolinium sublattice distinguished by an extremely diffuse type of  $\sigma_{s}(T)$ curve? Obviously the diffuseness, in the case of gadolinium ferrite-garnet, cannot be explained by fluctuations of the concentrations of magnetic and nonmagnetic ions over the volume of the ferrite, such as occur for example in the case of the alloy 30% Cu, 70% Ni.<sup>[5]</sup> The fact is that the ferritegarnets are distinguished by a strictly ordered distribution of ions in the lattice. In our opinion the diffuse form of the  $\sigma_s(T)$  curve for the gadolinium sublattice is explained by the fact that the establishment of long-range magnetic order in this sublattice depends on two interactions: the positive exchange interaction between  $Gd^{3+}$  and  $Gd^{3+}$  ions (in sublattice c) and the negative exchange interaction between Gd<sup>3+</sup> and Fe<sup>3+</sup> ions (action of the iron sublattice upon sublattice c).

The effect of the negative exchange interaction on the long-range magnetic order in sublattice c will be greater, the smaller the positive exchange interaction within sublattice c. Therefore if the positive exchange interaction is "switched off" on increase of temperature, the long-range magnetic order, though it decreases abruptly, does not completely disappear, since the negative exchange interaction delays its destruction. This also explains the diffuse character of the  $\sigma_{\rm S}(T)$ curve of the gadolinium sublattice. The negative exchange interaction in gadolinium ferrite is of appreciable magnitude, as is indicated by the comparatively high value of the compensation temperature (without negative exchange interaction between the sublattices, occurrence of a compensation point would be impossible).



Everything that has been said leads to a condition under which, with maximum probability, the occurrence of a compensation point in ferrites is to be expected. This condition is that in the ferrite one of the sublattices must have strong exchange interaction, and consequently a Weiss type of  $\sigma_{s}(T)$ curve (Fig. 2a, curve 1), and another sublattice must have "weak" exchange interaction, which leads (in the presence of antiferromagnetic interaction from the "strong" sublattice) to a spreadout temperature variation of  $\sigma_{s}(T)$  (Fig. 2a, curve 2). The compensation point  $\Theta_{C}$  occurs as a result of the subtraction from curve 2 (corresponding to the sublattice with the greater magnetic moment at 0°K) of curve 1 (with the smaller magnetic moment at 0°K). If the "weak" sublattice has a smaller magnetic moment at 0° K (Fig. 2b, curve 2) than has the "strong" (Fig. 2b, curve 1), then the subtraction of the curves results in an anomalous temperature variation of  $\sigma_{\rm S}({\rm T})$  of the ferrite at low temperatures (curve 3, curve type M or P of Néel<sup>[6]</sup>). Similar curves were observed in yttrium ferrite with part of the  $Y^{3+}$  ions replaced by ions  $Tb^{3+}$ ,  $Gd^{3+}$ , etc. As a special case, Néel's type L curve can be obtained if the magnetic moments of the "weak" and "strong" sublattices at 0° K are equal (curve 3 emerges from 0°K), i.e., the compensation point is located in the immediate vicinity of 0° K.

3. We now consider a lithium ferrite-chromite, for example of composition  $\text{Li}_{0.5}\text{Fe}_{2.5-a}\text{Cr}_aO_4$ (with a = 1.25), which according to Gorter<sup>[7]</sup> possesses a compensation point. In this ferrite there are two antiferromagnetically interacting sublattices; on the first of these are located Fe<sup>3+</sup>  $\text{Cr}^{3+}$ , and  $\text{Li}^{1+}$  ions (octahedral sites), on the second Fe<sup>3+</sup> and  $\text{Li}^{1+}$  ions (tetrahedral sites). At 0° K, according to a calculation by Néel's method, the magnetic moment of the first sublattice is equal to 5.45  $\mu_{\text{B}}$ , of the second 4.55  $\mu_{\text{B}}$ . In the low-temperature region, the ferromagnetism of the ferrite is determined by the octahedral sublattice. Indirect experimental evidence suggests that there is weaker exchange interaction in the octahedral sublattice than in the tetrahedral. Such evidence is provided by measurements of the magnetic properties of chromium ferrites, which, the measurements have shown, possess low Curie points. Furthermore, in the presence of antiferromagnetic interaction between the octahedral and tetrahedral sublattices, the former must have a spread-out type of  $\sigma_{\rm S}({\rm T})$ curve. Here also, therefore, conditions for the appearance of compensation points emerge.

We point out that according to Néel,<sup>[6]</sup> a  $\sigma_{\rm s}(T)$ curve with a compensation point can be obtained also from  $\sigma_{\rm sB}(T)$  and  $\sigma_{\rm sA}(T)$  curves (for the octahedral and tetrahedral sublattices) of the Weiss type, provided the sublattice magnetizations  $\sigma_{\rm sB}$  and  $\sigma_{\rm sA}$  (near 0°K) are nearly equal. Then in consequence of the presence of very small irregularities in the temperature variation on the Weiss curves (cf. Fig. 2c), appearance of a compensation point is possible here also. However, such a case is seldom realized in practice. Furthermore it follows from Fig. 2 that the compensation point can occur only at temperatures close to the Curie point, whereas in real ferrites it is usually observed at a distance from the Curie point.

4. Everything that has been said above leads to the interesting conclusion that in ferrites with a compensation temperature, there should occur a ''low-temperature'' point, corresponding to an abrupt change of the magnetic long-range order in the sublattice with the ''weak'' negative exchange interaction. This point should be accompanied by anomalies in the temperature variation of magnetic and other physical characteristics of the ferrite.

Ped'ko<sup>[2]</sup> has studied in detail, for gadolinium ferrite-garnet at liquid-nitrogen temperature, the anomalies of the temperature dependence of the paraprocess susceptibility, the magnetostriction, the coercive force, the width of the resonance

curve, and the resonance field; and he has shown that they are of a type reminiscent of those ordinarily observed at the Curie point. Goryaga and Lin Chang-ta,<sup>[4]</sup> in experiments with lithium ferrite-chromite of various compositions, showed that if a compensation point occurs in the ferrite, then there appear in it at the same time, in the low-temperature region, anomalies in the temperature variation of the spontaneous magnetization, coercive force, magnetic susceptibility, galvanometric effect, and electrical resistivity. The nature of these anomalies suggests that in the lowtemperature region a "Curie point," as it were, of the weak sublattice is manifesting itself. However, from the point of view of the thermodynamics of phase transitions one should, strictly speaking, not use the term "Curie point" here, since the change of long-range order in the "weak" sublattice comes about in the presence of the effective field of the "strong" sublattice.

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<sup>2</sup>A. V. Ped'ko, JETP **41**, 700 (1961), this issue p. 505.

<sup>3</sup>E. W. Lee and R. R. Birss, Proc. Phys. Soc. (London) **76**, 411 (1960).

<sup>4</sup>A. H. Goryaga and Lin Chang-ta, JETP 41, 696 (1961), this issue p. 502.

<sup>5</sup>K. P. Belov, Magnitnye prevrashcheniya (Magnetic Transformations), Fizmatgiz, 1959.

<sup>6</sup> L. Néel, Ann. phys. **3**, 137 (1948).

<sup>7</sup>E. W. Gorter, Philips Res. Reports **9**, 295 (1954).

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