Effect of a magnetic field on the nature of the Verve crystallographic transition in magnetite

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The effect of an external magnetic field on the nature of the Verve crystallographic transition in magnetite has been studied. A field imposed near this transition induces an incommensurable crystal structure in the magnetite.

Considerable theoretical and experimental effort has recently been devoted to the effects of external magnetic fields on the nature of various phase transitions (magnetic, spin-flip, crystallographic, etc.).[1–4]

Magnetite exhibits a Verve crystallographic transition near 119 K, where the space group changes from O_h^7 to C_{2v}^7. We were interested in determining whether a magnetic field would affect the nature of this transition.

As the test sample we chose a natural magnetite (Fe_3O_4) single crystal with a ratio $Fe_2O_3/FeO = 1.070$ containing, according to a chemical analysis, a small number of Ti^{4+} impurity ions. At 293 K this sample is a single-phase spinel with a lattice constant $a = 8.390$ Å, according to x-ray measurements. Using this sample we studied the effect of a magnetic field on the behavior of the thermal expansion $\Delta l/l$ along three crystallographic axes: [100], [110], and [111]. The measurements of $\Delta l/l$ were carried out with strain gauges in a bridge circuit. Curves of $\Delta l/l(T)$ were recorded on an x-y recorder. A $\Delta l/l(T)$ curve was first recorded over the temperature interval 80–160 K in the absence of an external magnetic field. The sample was then cooled at 3–4 deg/min to 80 K in a field $H = 6$ kOe; after this field was removed, a $\Delta l/l(T)$ curve was recorded again.

The dashed curves in Fig. 1 are $\Delta l/l(T)$ curves recorded at $H = 0$, while the solid curves are the $\Delta l/l(T)$ curves obtained after cooling in a magnetic field. Near the Verve transition the magnetic field has a strong effect on the thermal expansion is anisotropic at $H = 0$, for example, only a compression of the sample occurs along all three crystallographic axes after the sample is cooled in the magnetic field. Figure 2 shows the temperature dependence of the linear expansion coefficient $\alpha = (1/l)(\partial l/\partial T)$. We see that the magnetic field has an important effect on the temperature dependence of α along the [100] and [111] axes. After cooling in a field, a clearly defined negative λ anomaly appears in the coefficient α along the [100] axis, while along the [111] axis the λ anomaly in the coefficient α changes from positive to negative. Along the [110] axis, the λ anomaly in α remains negative but becomes considerably larger in magnitude.

In a theoretical paper[5] based on the theory of Landau phase transitions, Vitebskiĭ showed that a modulated crystal structure might arise by virtue of general symmetry considerations near a second-order crystallographic transition in magnetically ordered substances under the influence of an external magnetic field. In this case, long-period

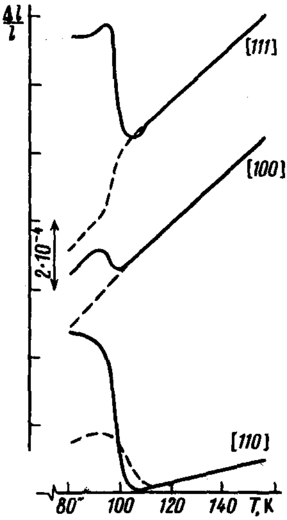

FIG. 1. Temperature dependence of the thermal expansion of a natural magnetite single crystal.

distortions with a period incommensurate with the period of the original lattice are superimposed on the original crystal structure. These modulated structures are called "incommensurate crystal structures." Transitions from a commensurate crystal structure to an incommensurate crystal structure are being studied by many investigators either by neutron scattering or by measurements of thermal expansion. According to

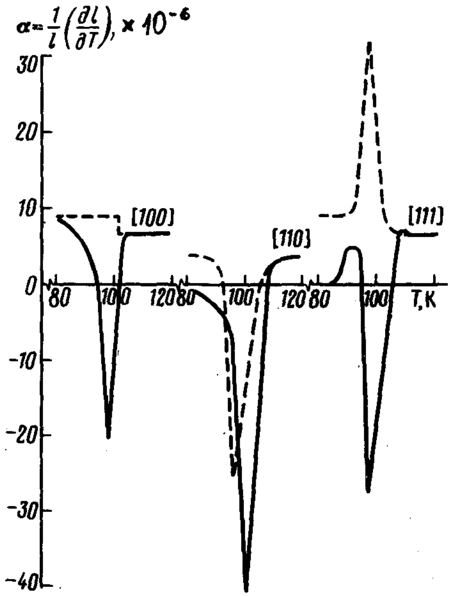

FIG. 2. Temperature dependence of the linear expansion coefficient α of a natural magnetite single crystal.

Refs. 6 and 7, a distinctive feature of an incommensurate crystal structure in a substance in the presence of a negative λ anomaly in the coefficient α near the crystallographic transition. Working from our results on the behavior of α as a function of the temperature and the conclusions of Refs. 5–7, we may assume that the imposition of an external magnetic field induces an incommensurate crystal structure near the Verve transition in magnetite.

This suggestion is supported by the results of neutron-scattering experiments.[8,9] Shirane *et al.*[8] and Fujii *et al.*[9] found that atoms are displaced distances on the order of $\Delta x = 0.02$ Å from their original positions in a magnetite sample cooled below the Verve transition temperature in a magnetic field.

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