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The Magnetic Properties of Ferroelectric and Ferromagnetic Ni-Cl Boracite at Low Temperature

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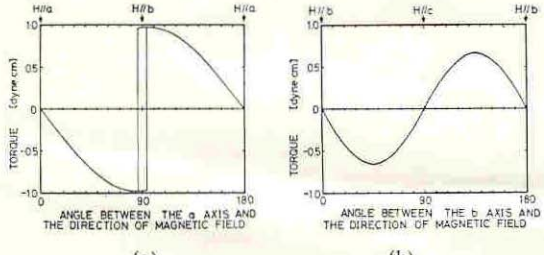
According to the previous measurements Ni-Cl boracite is weakly ferromagnetic below the Curie temperature (T_c) 15 K.[1] Recently, the magnetoelectric effect on this boracite composition has been measured.[2] The results show that the magnetoelectric susceptibility α_{32} has a temperature-dependence peak at 8.5 K, changes sign at 10.5 K, and becomes zero at about 25 K. From these results it has been suggested that the true Curie temperature of Ni-Cl boracite is 9 K, and that this crystal is antiferromagnetic between 9 K and 25 K. In this letter we report the results of magnetic torque measurements on a Ni-Cl boracite single crystal which corroborate this hypothesis.

The specimen was a thin rectangular single-crystal plate with its main surfaces parallel to the (001) plane and its edges along the [100] and [010] axes of the orthorhombic lattice. The size was $1.28\times1.56\times0.035$ mm³. Prior to measurement it had been polarized by application of an electric field of 26 kV/cm between the surfaces, which were covered with transparent gold electrodes. The fact that the specimen was in a ferroelectric single-domain state was verified by observation under polarized light. The magnetic torque was measured with an automatic recording torquemeter between 4.2 K and 77 K in a magnetic field up to 6700 Oe. The direction of magnetic field



(a) (b)

Fig. 1. The torque curves of a Ni-Cl boracite single crystal (0.80 mg) in the magnetic field of 1700 Oe at 4.2 K. (a) Magnetic field in the *a-b* plane. (b) Magnetic field in the *b-c* plane.

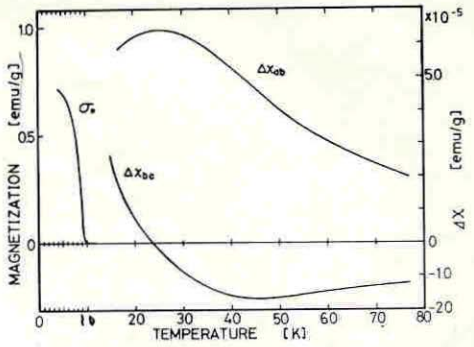


Fig. 2. Temperature dependence of the magnetization and the differences in susceptibility in Ni-Cl boracite (obtained from the magnetic torque).

was rotated in the three orthorhombic principal planes. The temperature was measured with a calibrated Au/Co-Cu thermojunction placed close to the specimen.

The torque curves at 4.2 K are shown in Fig. 1. The angular dependence in the *a-b* plane (Fig. 1 (a)) has a sharp jump, which is proportional to the applied magnetic field, when the magnetic field becomes parallel to the *b* direction. This behavior is typical of a weak ferromagnet. From the size of the jump we get the spontaneous magnetization σ_0 at 4.2 K of 0.72 emu/g, which is about twice as large as the previously reported value measured on a powder specimen.[1] As the temperature rises this dependence continuously turns into an ordinary twofold type and the magnitude of the maximum torque decreases rapidly (Fig. 2) towards 9.7 K. Above this temperature the torque curves are entirely of twofold type with easy direction along the *a* axis. Their amplitudes are proportional to the square of the applied magnetic field and show a broad peak near 25 K (Fig. 2). On the while the angular dependence in the *b-c* plane is of an ordinary twofold type (Fig. 1 (b)), and its maximum value is proportional to the square of the applied magnetic field over the whole temperature range. With rising temperature the easy direction changes from the *b* to the *c* direction at 25 K.

These experimental results are easily understood if Ni-Cl boracite is a weak ferromagnet below 9.7 K and a collinear antiferromagnet with the sublattice moments directed in the *c* direction between 9.7 K and about 25 K.

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