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Reference


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POLARIZED LIGHT STUDIES OF FERROMAGNETIC/FERROELECTRIC/FERROELASTIC DOMAIN PATTERNS IN NiCl and NiBr BORACITE

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Abstract - Ferromagnetic stripe domains have been observed in polarized light along the optic axes of the orthorhombic NiCl and NiBr boracites below ~9K and ~30K, respectively. The spontaneous magnetization vector is found to lie along the principal refractive index nγ. Measurements of spontaneous Faraday rotation and spontaneous birefringence down to 4.2K are presented.

INTRODUCTION

The 3d-metal boracites M3B7O13X, abbreviated MX, with the compositions M=Fe, Co, Ni, Cu and X=Cl, Br, I (except CuI) are known to be weakly ferromagnetic ferroelastic ferroelectrics, being transparent in the visible. So far observations of ferromagnetic domains have been reported for NiI3,4,5 and CoCl2 only. In this work the compositions remaining crystallographically orthorhombic down to 4.2K, MnCl,MnBr,MnI,NiCl,NiBr, CuCl and CuBr, have been examined with a view to make visible ferromagnetic domains. For NiCl and NiBr the search was successful; their respective spontaneous magnetizations Ms at 4.2K were found to be 0.72 emu/g (single crystal)6 and 2.15 emu/g (powder)1. Magnetoelectric7 and magnetic torque6 measurements on NiCl yielded a magnetic Curie point of ~10K.

SPONTANEOUS BIREFRINGENCE

In order to make visible ferromagnetic domains in strongly birefringent NiCl and NiBr, one has to observe along or close to an optic axis. This is due to strong interference of birefringent phase retardation with Faraday rotation. Orthorhombic NiCl and NiBr are optically positive with nγ//P/\c|b0,r.\|/\<001>cub, n0//b0,r.\|/\<110>cub and nγ//a0.r.\|/\<110>cub with the optic axes lying in the (001)0.r. plane. With a view to locating the optic axes of NiCl and NiBr in the ferromagnetic range, we have extended the measurements of the principal birefringences of NiCl and NiBr from room temperature to 4.2K. (see Figs. 1a and b). Using Mallard's approximation formula10, sinVγ=[(nγ-nα)/(nγ-nβ)]1/2, where Vγ is half the acute optic angle and nγ-nα, nγ-nβ are principal birefringences, the optic angles 2Vγ of ~79° and ~71° are obtained for NiCl and NiBr at 4.2K, respectively. This means that on (100)cub cut platelets with spontaneous polarization within that plane (Fig.2a) one of the two optic axes is coming out at ~5.5° and ~9.5° off the vertical for NiCl and NiBr, respectively. As shown below, these angles and the accompanying weak birefringence nγ-nβ proved acceptable for observation of magnetic domains (compare Figs 4 and 5). For NiCl also the birefringence section nγ-nβ has been measured between 4.2 and 40K (Fig. 3) using a lock-in amplifier together with a photoelastic modulator11. Below 10K a contribution from the Faraday rotation is clearly observed.

FERROMAGNETIC DOMAINS AND ORIENTATION OF SPONTANEOUS MAGNETIZATION VECTOR

For observing ferromagnetic domains in the interior of the ferroelectric ferroelastic domains,(100)cub-cut platelets of NiCl and NiBr were used, allowing in principle,
the simultaneous observation of four different ferroelectric domains having $P_S$ within the plane (see Table 2 of ref.4). At nearly crossed polars, close to extinction, ferromagnetic domains have been observed in NiCl (Fig.4) and NiBr (Fig.5) in the interior of such ferroelectric domains. In white light brown/bluish contrast was obtained between different states of magnetization. Minimization of magnetostatic energy leads to narrow stripe domains after cooling in the field free state. The stripes are separated by walls following approximately (001) orth. rh. planes. The NiBr sample (Fig.5) is particularly instructive because it shows ferroelectric domains with four different orientations of $P_S$ and all three types of ferroelectric/ferroelastic domain walls known for NiCl and NiBr 4, i.e. "180°", "head-head(tail-tail)" and "head-tail" (Fig.6).

With a view to deducing the orientation of the spontaneous magnetization vector $M_s$ in NiCl and NiBr, stability tests of the stripe patterns in an external field $H$ (~300G, permanent magnet rotated around the cryostat) were performed: (i) for rotation of $H$ within the plane $n_S/n_\alpha$, the stripes are stable for any angular position of $H$; (ii) for rotation of $H$ within the plane $n_S/n_\alpha$, the stripes are stable for $H$ parallel to $n_S$ only; (iii) for $H$ rotating within the plane $n_\gamma/n_\alpha$, the stripes are stable for $H$ parallel to $n_\alpha$ only. From (i) to (iii) $M_s$ is found to lie parallel to $n_\gamma$ (a_0-axis of ref. 7) in good agreement with results from magnetoelectric and torque measurements on NiCl.

From this result the schematic representation of the stripe domains follows (Fig.2b) and which - together with Fig.2a-explains the magnetic domain patterns of Figs.4 and 5. The angles between $M_s$ on either side of the ferroelectric walls 1, 2 and 3 (Fig.2) are 90°, 60° and 60°, respectively. Whereas across the walls 2 the magnetic domains seem to be joined both by magnetic head-head and head-tail junctions, the continuity of the stripes across the walls 3 seems to indicate that in the latter case the magnetic stripes - while "bending" around 90° - are joined by head-head junctions.

**FARADAY ROTATION**

In Figure 6 the apparent Faraday rotation versus temperature of NiCl and NiBr is shown for observation perpendicularly to (100) cubcuts containing $P_S$ within the plane and characterized by the birefringence $n_\gamma-n_\alpha$. By 'apparent' we mean that the plotted rotation is the angle formed by the plane of the initial linearly polarized wave and the major axis of the elliptically polarized wave leaving the crystal. For measurement a conventional technique using a polarizer, an analyser and a microphotometer was used in conjunction with a photoelastic modulator acting as a high frequency polarization chopper.

**CONCLUSIONS**

The domain studies of NiCl and NiBr lead to the deduction of Aizu species 43m1 'Fm'm2' for both compositions at 4.2K, with $M_s$ lying in both cases along $n_\gamma$. Further work is needed for separating the intrinsic Faraday rotation from birefringent phase retardation.

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FIGURE 1 Principal spontaneous birefringences versus temperature, (a) NiCl, (b) NiBr ($\lambda = 546\,\text{nm}$; tilting compensator)

FIGURE 2 Schematic domains on a (100)$_{\text{cut}}$ cut (a) ferroelectric/ferroelastic single domain, (b) ferromagnetic stripe domains occurring in (a)

FIGURE 3 Spontaneous birefringence $n_y - n_x$ of NiCl versus temperature (photoelastic modulator). Sample thickness, 244$\mu\text{m}$

FIGURE 4 Ferromagnetic stripe domains of NiCl, (a) narrow stripes, as-cooled without magnetic field; $T = 4.2\,\text{K}$, (b) large stripes after repeated switching; $T = 4.2\,\text{K}$. Sample thickness, 244$\mu\text{m}$.
FIGURE 5 Ferromagnetic stripe domains of NiBr, (a) as-cooled without field, T = 12K, (b) ferroelectric/ferroelastic domains of Fig. 5a; 1, 2, 3, are 180°, head-head and head-tail domain walls, resp. (compare Fig. 6). Sample thickness, 77μm.

FIGURE 6 Three types of ferroelectric/ferroelastic domain wall occurring in NiCl and NiBr.

FIGURE 7 Apparent Faraday rotation of NiCl and NiBr; λ = 546nm; sample thicknesses 244μm and 145μm, resp.

REFERENCES