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Influence of Tetrahedral  $\text{Ni}^{2+}$  Ions on the Magnetic Properties  
of YIG

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In a previous paper (1) we have shown by studying the anisotropic energies and the lattice constants of NiO doped yttrium iron garnet single crystals ( $\text{GeO}_2$  being used for charge compensation), that  $\text{Ni}^{2+}$  ions enter the tetrahedral (24d) sites although in a very small quantity. It is known that  $\text{Ni}^{2+}$  ions ( $d^8$ ) cause Jahn-Teller distortions of the oxygen tetrahedra, as for example in nickel ferrite chromites (2). However, when its concentration is very low as in our case, one could expect the distortions to occur only locally, though the structure as a whole could still conserve the cubic symmetry. This type of distortion is known as dynamic Jahn-Teller distortion.

The deformations being oriented at random, one can not expect them to modify the cubic nature of the overall anisotropy as measured by the usual methods, though such deformations do seem to influence the line width (3). In order to verify the above hypotheses in our materials, we have studied in this note: 1. the angular dependence of the field for resonance  $H_0$ , in order to see if it could be described adequately by the two constants  $K_1$  and  $K_2$  of cubic anisotropy and 2. the line width and its temperature dependence.

Two compositions of doped YIG containing 0.25 and 1.3 weight% NiO were investigated. Spheres used for the measurements were of about 0.4 mm diameter polished to 5  $\mu\text{m}$ . They were oriented by the usual X-ray techniques such that  $H_{\text{DC}}$  could be applied in the (110) plane and they could be rotated about the [110] axis. The measuring frequency was about 9100 MHz and the temperature range of investigations

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Table 1

	NiO 0.25%		NiO 1.3%	
	20 °C	-190 °C	20 °C	-190 °C
$\frac{K_1}{M}$	-19	+190	-12	+375
$\frac{K_2}{M}$	-	-460	-	-690

was from -190 to +60 °C.

Angular Dependence of  $H_0$  By applying Kittel's equation for resonance to crystals of cubic symmetry one can write:

$$\left(\frac{\omega}{\gamma_{\text{eff}}}\right)^2 = \left[ H_0 + \frac{K_1}{M} (2 - \sin^2\theta - 3\sin^2 2\theta) + \frac{K_2}{2M} \sin^2\theta (6\cos^4\theta - 11\sin^2\theta\cos^2\theta + \sin^4\theta) \right] \times$$

$$\times \left[ H_0 + \frac{K_1}{M} (2 - 4\sin^2\theta - \frac{3}{4}\sin^2 2\theta) - \frac{K_2}{2M} \sin^2\theta\cos^2\theta (3\sin^2\theta + 2) \right],$$

where  $\omega$  is the measuring frequency,  $\gamma_{\text{eff}}$  the effective gyro-magnetic ratio (given by  $ge/2mc$ , where  $g$  is the spectroscopic splitting factor),  $H_0$  the field for resonance,  $K_1$ ,  $K_2$  the two constants of anisotropy,  $M$  the saturation magnetization and  $\theta$  the angle between  $H_0$  and  $[001]$  axis in the  $(110)$  plane.

From experimentally determined values of  $H_0$  along  $[001]$ ,  $[110]$ , and  $[111]$  directions at -190 and +20 °C,  $K_1/M$  and  $K_2/M$  were calculated for the above temperatures (Table 1) and these values in turn were used to compute the angular dependence of  $H_0$  both at -190 and +20 °C. These curves were compared with those obtained experimentally. Fig. 1 shows the results obtained at -190 °C for both compositions (the results at 20 °C are not shown since  $K_1$ ,  $K_2$  being quite low the angular dependence of  $H_0$  is less remarkable), and the agreement seems to be satisfactory. This indicates that the two cubic constants  $K_1$  and  $K_2$  are quite adequate to describe the anisotropic energy. So the dynamic Jahn-Teller distortions do not seem to perturb the cubic symmetry of the anisotropy as we could expect it.

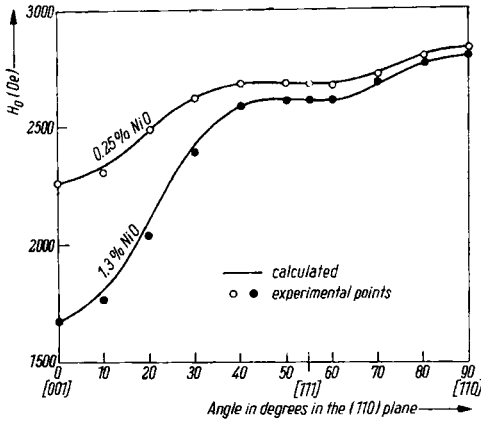


Fig. 1

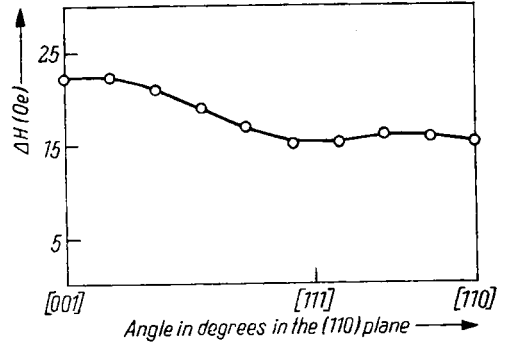


Fig. 2

**Line Width  $\Delta H$**  The angular dependence of  $\Delta H$  studied in these materials shows an anisotropic behaviour with a maximum along the  $[001]$  direction. Fig. 2 shows the result for YIG + 0.25% NiO. The line width also increases with the NiO content.

As the temperature is decreased,  $\Delta H$  increases, this being most pronounced along the  $[100]$  direction as shown in Fig. 3. The variation of  $\Delta H$  along the  $[110]$  direction, being similar to that along  $[111]$ , is omitted in the figure for clarity.

This increase in line width can not be attributed to non-saturation effects, since  $H_0$  is still higher than the demagnetizing field  $4\pi M/3$  and further the  $[100]$  direction actually becomes the easy axis at lower temperatures (1). It is interesting to remark that a similar behaviour of  $\Delta H$  was observed in the case of YIG containing  $Mn^{3+}$  ions, which was supposed to be caused by the dynamic Jahn-Teller distortions of the oxygen octahedra occupied by  $Mn^{3+}$  ions (3). The rf field is considered to be coupled to the distortion, by either bulk magneto-strictive effects or the local anisotropy of the Jahn-Teller site. In our case, this increase may

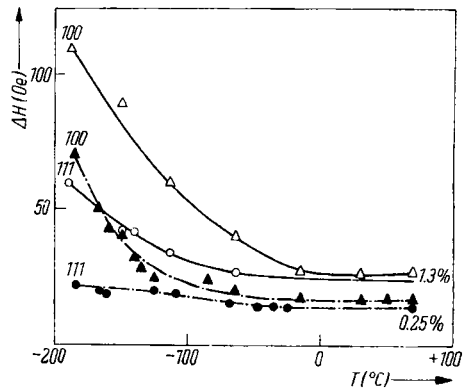


Fig. 3

be attributed in the same way to the dynamic distortions of the oxygen tetrahedra occupied by  $\text{Ni}^{2+}$  ions. It is seen that the increase in  $\Delta H$  say at  $-190^\circ\text{C}$  over its value at  $20^\circ\text{C}$ , in the case of  $\text{Ni}^{2+}$  ions is smaller than it is for  $\text{Mn}^{3+}$ . This may be due to the actual concentration of tetrahedral  $\text{Ni}^{2+}$  ions which is extremely low.

Jahn-Teller distortions of the dynamic type seem to be caused by the tetrahedral  $\text{Ni}^{2+}$  ions in YIG, as shown by the low temperature increase of the line width. The angular dependence of  $\Delta H$  is also found to be anisotropic. Such distortions however do not seem to perturb the global cubic anisotropic energy.

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