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# Antiferromagnetic Magnetostriction in CoO and NiO Single Crystals

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The magnetostriction in CoO and NiO single crystals was measured along the principal crystallographic directions at room and liquid air temperatures. It has been confirmed, from the measurement of the temperature dependence of the magnetostriction in CoO crystals, that the magnetostriction vanishes near the Néel temperature. It has been found that the magnetostriction,  $\lambda$ , depends on the direction,  $\phi$ , and the intensity, H, of the applied magnetic field, as  $\lambda = [a+b\cos 2(\phi-\phi_0)]H^2$ , where a, b and  $\phi_0$  or the direction of the field where the magnetostriction is maximum, are constants.

#### Introduction

In connection with the nature of the lattice deformation accompanied with the magnetic transformation in antiferromagnets,<sup>1)</sup> it may be important to investigate the nature and behavior of magnetostriction in those substances, which was previously predicted to be observed.<sup>2)</sup> Recently, Belov and Levitin<sup>3)</sup> reported briefly some magnetostrictive behavior of NiO polycrystals measured in magnetic fields up to 14,000 Oe. To get more detailed information about the antiferromagnetic magnetostriction, especially about its anisotropic behavior, we made the magnetostriction measurements on single crystals of CoO and NiO.

## Crystal specimens and the method of measurements

The crystal specimens used are two circular plates with plate surfaces parallel to an (100) plane, 10 mm in diameter and 1 mm in thickness (commonly to CoO and NiO), and two rectangular plates with the plate surfaces parallel to an (110) plane and the dimension  $7 \times 5 \times 0.7 \text{ mm}^3$  (NiO) and  $5 \times 4 \times 0.5 \text{ mm}^3$  (CoO). These four specimens were cut out, making use of the cleavage and using a diamond cutter from four block single crystals grown by Dr. Y. Nakazumi of Fuji Titanium Industrial Co. using the Verneuil method. They were polished with Emery papers 0 5, and etched with boiling aqua regia for NiO or with hydrofluoric acid for CoO for the determination of crystal orientations by the light figure method,<sup>4)</sup> and then, without any heattreatment, submitted to the magnetostriction

measurements, which were performed by the strain gauge technique<sup>5)</sup> at room and liquid air temperatures in magnetic fields up to 11,000 Oe. Longitudinal and transverse magnetostrictions were measured along the [001] and [0Ī1] directions in (100) crystal specimens and along the [001], [1Ī0], and [1Ī1] directions in (110) crystal specimens.

### **Experimental results**

In CoO crystals, the magnetostriction could not be detected at room temperature but a fairly large magnetostriction was found at liquid air temperature. While, in NiO crystals, the magnetostriction was observed both at room and liquid air temperatures.

From the measurement of the variation of magnetostriction with temperature in CoO crystals, it has been confirmed that the magnetostriction decreases gradually with in-



Fig. 1. Temperature dependence of the longitudinal and transverse magnetostrictions measured along the [001] direction in the (100) disc of CoO single crystal.



Fig. 2. Variations, with the intensity, H, (a) and direction,  $\phi$ , (b) of the applied field, of the longitudinal (triangles) and transverse (reversed triangles) magetostrictions measured along the [001] direction in the (100) disc of CoO single crystal at liquid air temperature. Circles in the figure (b) represent the data of magnetostriction measured for every 10 degree rotation of magnetic field of 10.8 kOe applied parallel to the disc surface, where  $\phi$  is the angle between the direction of the field and the [001] direction. Solid lines are calculated from the relation  $\lambda = [a+b\cos 2(\phi-\phi_o)]H^2$ .

creasing temperature and vanishes near its Néel temperature (Fig. 1), as found previously with NiO polycrystals by Belov and Levitin<sup>2)</sup>. Thus, we can say that the magnetostriction observed here is originated from antiferromagnetism.

Both the longitudinal and transverse magnetostrictions measured along the abovementioned directions vary in proportion to the square of the intensity of magnetic field, as shown in Fig. 2(a). This is contrasted to a linear increase of the magnetostriction beyond a distinct critical field as claimed by Belov and Levitin<sup>2)</sup> with NiO polycrystals. Also, the variation of the magnetostriction observed when an applied magnetic field of a constant intensity is turned around the axis normal to the specimen surface is of the  $\cos 2\phi$  type, where  $\phi$  is the angle between the direction of the field and a certain standard direction (actually the [001] direction was taken as the standard direction) in the plate surface of the specimen, as shown in Fig. 2(b). These two features are common to the antiferromagnetic magnetostriction observed along any direction of measurement in CoO and NiO single crystals. Consequently, we can express the relation between the measured magnetostriction,  $\lambda$ , and the intensity, H, and direction,  $\phi$ , of the applied field as

 $\lambda = [a + b \cos 2(\phi - \phi_0)] H^2 , \qquad (1)$ 

where a and b are arbitrary constants deter-

mined so as to fit the measuring points, and  $\phi_0$  represents the direction of the applied field where the maximum magnetostriction is observed. It has been found that the values of a and b in Eq. (1), namely the values of the level and amplitude of the  $\lambda - \phi$  curve vary to a considerable extent by replacing the strain gauge for measurement by a new one, as, for example, shown in Fig. 3, but the value of  $\phi_0$  does not change so much as shown in Table 1. Hence,  $\phi_0$  may be regarded as an indicator of the anisotropic behavior of the antiferromagnetic magnetostriction. Some examples of the observed



Fig. 3. Variation of the  $\lambda$ - $\phi$  curve observed at room temperature, when the strain gauge set along the [011] direction in the (100) disc of NiO single crystal was replaced by a new one.

Table I. The observed data on the direction of the field,  $\phi_0$ , where the magnetostriction,  $\lambda$ , is maximum, in the magnetostriction measurements along the principal crystallographic directions in single crystals of CoO and NiO.

Specimen	Direction of Measurement	<i>\$</i> 0
(100) CoO	[001] [011]	$5^{\circ} \sim 10^{\circ} ([001])$ 120°~145° ([011])
(110) CoO	[001] [1Ī0] [1Ī1]	$\begin{array}{c ccccc} -15^{\circ} \sim & 0^{\circ} & ([001]) \\ 75^{\circ} \sim 110^{\circ} & ([1\overline{1}0]) \\ 10^{\circ} \sim & 35^{\circ} & ([001] \sim [1\overline{1}2]) \end{array}$
(100) NiO	[001] [011]	$\begin{array}{c} -10^{\circ} \sim 20^{\circ} \ ([001]) \\ 25^{\circ} \sim 40^{\circ} \ ([0\overline{1}1]) \end{array}$
(110) NiO	[001] [1Ī0] [1Ī1]	$\begin{array}{c} 0^{\circ} & ([001]) \\ 70^{\circ} \sim 85^{\circ} & ([1\overline{10}]) \\ 100^{\circ} \sim 125^{\circ} & ([1\overline{10}] \sim [1\overline{11}]) \end{array}$



Fig. 4. Anisotropic behavior of the magnetostriction measured along the principal crystallographic directions in single crystals of CoO and NiO.

 $\lambda - \phi$  curves are shown in Fig. 4. As may be seen from this figure, the magnetostriction in CoO crystals is larger than that in NiO crystals. It is to be noticed, further, that the  $\phi_0$  values observed in the measurements along the [011] and [111] directions are different by 90° for CoO and NiO single crystals, as seen from Table I.

The experimental results obtained with CoO single crystals can be understood well in terms of the concept of the domain wall displacements in antiferromagnets as proposed by Néel<sup>6)</sup> by taking into considerations the data of the lattice deformation and the direction of spin as determined by X-ray<sup>1)</sup> and neutron diffraction measurements<sup>7)</sup>. But, the observed behavior of antiferromagnetic magnetostriction in NiO single crystals is still unclarified because of the lack of information about the magnetic anisotropy in the (111) plane.

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### DISCUSSION

S. IIDA: In a previous study by U.S.S.R., the presence of a critical field was reported for the magnetostriction of the antiferromagnets, contrary to your experiments. Could you explain the reason of this discrepancy?

T. NAKAMICHI: The magnitude of magnetostriction obtained by Belov and Levitin is very small. In such a small magnetostriction, it might be very difficult to determine a precise field dependence of magnetostriction. So, we cannot agree with their assertion of a linear increase of magnetostriction beyond a certain critical field.

However, if the potential energy of the domain wall has a special form in their sintered polycrystalline NiO specimen, the presence of such a critical field might be expected, which was not found in our single crystal specimens.

S. IIDA: Mr. K. Kon of our laboratory has made a measurement of tilt angles of NiO single crystal by using finely line focussed monochromatic X-ray. He has found a presence of a tilt angle of about 1 minute which can be affected by an applied magnetic field. This may correspond to the antiferromagnetic domain boundary of a single twin domain of this rhombohedral antiferromagnet.

Y. SHIMOMURA: I believe that your single crystal specimens must certainly be twinned. I am afraid that the results of observation would vary with the distribution of twin components.

T. NAKAMICHI: Yes, I agree with your comments. But, it is difficult to observe the magnetostrictive behavior in the single domain crystal which has no twin structure, using the strain gauge, except for a special case. We are now trying to observe magnetostriction in (111) surface specimen of NiO single crystal, in which it is possible to maintain a single domain when a strain gauge is cemented on that crystal surface.