

$3d^8(\text{cryst})$  states is rather small, the  $3d^8(\text{cryst})$  states have spread in energy as a consequence of the disorder in the film, and the background photocurrent in the quasicontinuum "band" is rapidly increasing with photon energy and may disguise the contribution from the  $3d^8(\text{cryst})$  states. In addition one may also speculate on an inherent reason: The  $3d^8(\text{spin})$  and  $3d^8(\text{cryst})$  excited states are of entirely different origin: While the former depends on a spin-spin interaction between several Ni atoms, the latter states can be interpreted as "internal" excitations of the  $\text{Ni}^{++}$  ion and hence may be less likely to be coupled to conduction-supporting states in the NiO system.

In conclusion, we believe that in disordered NiO there exists a coupling between nonconducting states and conducting states. As a result, the spectral response of the photoconductivity shows a structure due to the nonconducting states. These measurements can be easily extended to other disordered systems in order to

obtain information on nonconducting states.

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#### LATTICE ANISOTROPY IN ANTIFERROMAGNETIC CHROMIUM\*

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Thermal expansion and magnetostriction measurements on single- $\vec{Q}$  single-crystal chromium give the lattice parameter ratios in the orthorhombic phase below the Néel temperature and through the transition to the tetragonal phase at the spin-flip temperature,  $T_F = 122^\circ\text{K}$ . The  $\vec{Q}$  direction is the longest axis at  $300^\circ\text{K}$  and becomes the shortest axis below  $\sim 200^\circ\text{K}$ . The anisotropic discontinuities in length at  $T_F$  result in a net volume dilatation,  $1.4 \times 10^{-6}$  with decreasing temperature.

Recent experimental work<sup>1,2</sup> has demonstrated the expected and long-sought<sup>3-5</sup> deviations from cubic symmetry of the lattice parameters of chromium in its antiferromagnetic state. In this paper we wish to report our measurements of these deviations over the temperature range from  $115^\circ\text{K}$  to the Néel temperature and show that the longest axis at  $300^\circ\text{K}$  becomes the shortest axis below  $220^\circ\text{K}$ .

At the Néel temperature,  $T_N = 312^\circ\text{K}$ , Cr undergoes a first-order transition<sup>6</sup> from a body-centered cubic paramagnetic state to an antiferromagnetic transverse spin-density-wave state ( $\text{AF}_1$ ). At the spin-flip temperature,  $T_F = 122^\circ\text{K}$ ,

Cr undergoes a first-order transition<sup>7</sup> from the transverse spin-density-wave state to one with longitudinal polarization ( $\text{AF}_2$ ). The wave vector of the spin density wave is denoted by  $\vec{Q}$  and the spin polarization by  $\vec{s}$ . Both  $\vec{Q}$  and  $\vec{s}$  lie along  $\langle 100 \rangle$  directions.<sup>4</sup> If a crystal of Cr is cooled through  $T_N$  in the presence of a sufficiently large magnetic field along a  $\langle 100 \rangle$  direction,<sup>8</sup> it will have a single  $\vec{Q}$  throughout most of its volume, with  $\vec{Q}$  parallel to the cooling field,  $\vec{H}_C$ . The field may be removed below  $T_N$  without disturbing the single- $\vec{Q}$  state. Graebner and Marcus<sup>9</sup> showed that a crystal cooled through  $T_N$  in zero field consists of regions in each of which there

is only one  $\vec{Q}$ . In the  $AF_1$  phase in a single- $\vec{Q}$  crystal in the absence of an applied field the spins may lie along either of the  $\langle 100 \rangle$  directions normal to  $\vec{Q}$ . However, when a large field is applied to a single- $\vec{Q}$  crystal below  $T_N$  but above  $T_F$  in a  $\langle 100 \rangle$  direction normal to  $\vec{Q}$ , the spins align themselves along the  $\langle 100 \rangle$  direction normal to both  $\vec{Q}$  and  $\vec{H}$ .<sup>4</sup> The orthorhombic symmetry of the  $AF_1$  phase is then apparent, with the three directions labeled  $\vec{Q}$ ,  $\vec{s}$ , and  $\vec{n}$ , as shown in Fig. 1. Below  $T_F$  the spins lie parallel to  $\vec{Q}$ , resulting in tetragonal symmetry with the directions labeled  $\vec{Q}$  and  $\vec{n}$ . We denote the lattice parameters in the  $AF_1$  phase in the  $\vec{Q}$ ,  $\vec{s}$ , and  $\vec{n}$  directions by  $c_1$ ,  $b_1$ , and  $a_1$ , respectively, and in the  $AF_2$  phase in the  $\vec{Q}$  and  $\vec{n}$  directions by  $c_2$  and  $a_2$ , respectively.

The application of a large field parallel to  $\vec{Q}$  depresses  $T_F$ ,<sup>7</sup> but if a sufficiently large field parallel to  $\vec{Q}$  is applied isothermally at a temperature below the depressed  $T_F$ , irreversible " $\vec{Q}$  switching" will occur<sup>10,11</sup> so that  $\vec{Q}$ 's normal to the original single  $\vec{Q}$  appear, allowing the spins (and thus  $\vec{Q}$ ) to lie in the energetically favorable position normal to the applied field. Information obtained from "stress-cooling" experiments<sup>11-14</sup> (the preferred direction for  $\vec{Q}$  is parallel to a tensile stress applied along  $\langle 100 \rangle$  while cooling

through  $T_N$ ) and thermal expansion measurements near  $T_N$ <sup>2</sup> indicates that near  $T_N$  the  $\vec{Q}$  direction is the longest direction in the orthorhombic unit cell. This means that  $c_1/a_1-1$  is positive near  $T_N$ , as is  $c_1/b_1-1$ . We have measured  $c/a-1$  and  $b/a-1$  as a function of temperature between 115°K and  $T_N$  and find that in addition to the abrupt changes in the magnitude of these quantities at  $T_F$ ,<sup>1</sup>  $c/a-1$  changes sign at  $\sim 230^\circ\text{K}$ . We have confirmed our determination of the sign of  $c_2/a_2-1$  at 100°K in a  $\vec{Q}$ -switching experiment.

We have carried out differential thermal expansion and magnetostriction measurements on a single- $\vec{Q}$  crystal of Cr using a three-terminal capacitance technique.<sup>15</sup> The single-crystal sample was spark-planed from an arc-melted boule<sup>16</sup> of volume  $\sim 1 \text{ cm}^3$  with two parallel faces normal to the cube axis denoted by  $Z$ . The other cube axes are denoted by  $X$  and  $Y$ . Measurements of length were always made in the  $Z$  direction. Cooling fields of 50 and 100 kOe were found to produce the same size discontinuities at  $T_F$ , from which we conclude that the cooling field of 100 kOe used throughout the work reported here, except as noted in the  $\vec{Q}$ -switching experiments, produces a single- $\vec{Q}$  state. The capacitance cell was made of a commercial molybdenum alloy,<sup>17</sup> which has a thermal expansion nearly equal to

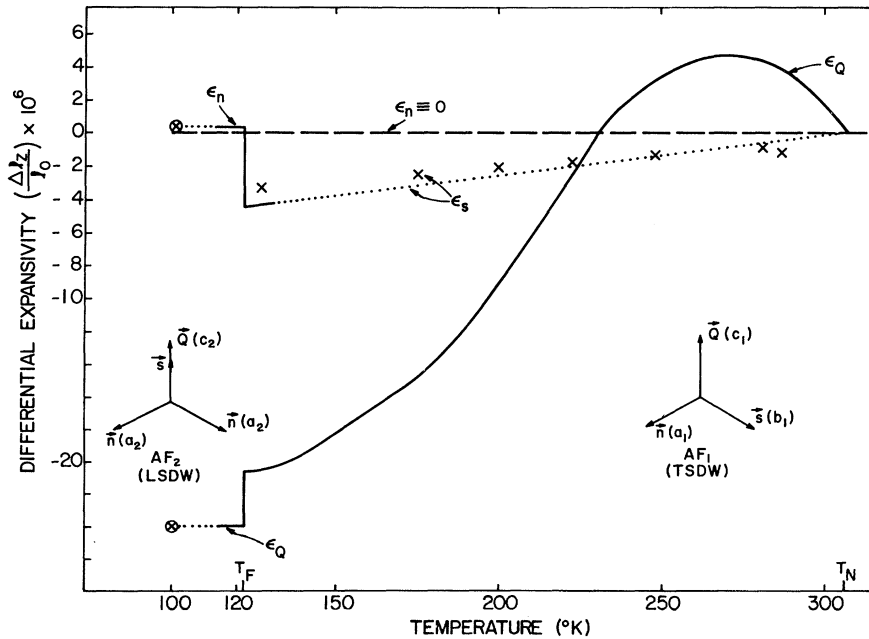


FIG. 1. Temperature dependence of differential thermal expansivity of single- $\vec{Q}$ , single-crystal chromium. Solid curves, crosses, and circled crosses represent data. The dotted curve is a linear interpolation of  $\epsilon_s$  between zero at  $T_N$  and  $\epsilon_s$  just above  $T_F$ . The dashed curve is our zero reference resulting from setting  $\epsilon_n \equiv 0$  in the  $AF_1$  phase. In the  $AF_1$  phase  $\epsilon_n \equiv 0$ ,  $\epsilon_Q = c_1/a_1 - 1$ ,  $\epsilon_s = b_1/a_1 - 1$ . In the  $AF_2$  phase  $\epsilon_n = a_2/a_1 - 1$ ,  $\epsilon_Q = c_2/a_1 - 1$ .

that of Cr. A 100-kOe superconducting solenoid was used for field cooling along the  $X$ ,  $Y$ , and  $Z$  directions, and for measurements requiring a field in the  $Z$  direction. A 22-kOe rotatable iron electromagnet was used for measurements with  $\vec{H}$  in the  $X$ - $Y$  plane. The state of the crystal with  $\vec{H}$  along  $X$  or  $Y$  or  $Z$  is specified by  $(\vec{Q}_i, \vec{s}_j)$ , where  $i = X, Y, Z$  and  $j = X, Y, Z$  specify the wave vector and spin directions.

Continuous recordings of the thermal expansion with respect to the cell were made as a function of temperature from below  $T_F$  to above  $T_N$  for increasing temperature only, as the single- $\vec{Q}$  state is destroyed on passing through  $T_N$ . In reducing the data, we make the assumption that Cr is cubic at  $T_N$ , and that all length changes can be referred to this state.<sup>18</sup> Since we measure differential thermal expansion we plot the difference in expansivity  $\Delta l_Z/l_0$  normalized to the  $\vec{n}$  direction in the  $AF_1$  phase. A temperature sweep from below  $T_F$  to above  $T_N$  was made for  $\vec{Q}$  along  $Z$ , in zero applied field, so that the spins could lie along either  $X$  or  $Y$  in the  $AF_1$  state denoted by  $(\vec{Q}_Z, \vec{s}_{X,Y})$ . This was compared with a sweep for  $\vec{n}$  along  $Z$  with  $\vec{Q}$  along  $Y$  and a field  $\vec{H}$  applied along  $Z$  so that all the spins lay along  $X$  in the  $AF_1$  phase  $(\vec{Q}_Y, \vec{s}_X)$ . The difference between these sweeps yielded the curve  $\epsilon_0$  in Fig. 1. Temperature sweeps were also made for increasing and decreasing temperature in a range of about 20°K around  $T_F$  to explore in more detail the discontinuous changes in length which occur at  $T_F$ . These sweeps were made for  $\vec{Q}$ ,  $\vec{s}$ , and  $\vec{n}$  in the  $Z$  direction, the latter two for  $\vec{Q}$  in first the  $X$  and then the  $Y$  direction—i.e., for  $AF_1$  states denoted by  $(\vec{Q}_Z, \vec{s}_{X,Y})$ ,  $(\vec{Q}_X, \vec{s}_Z)$ ,  $(\vec{Q}_Y, \vec{s}_Z)$ ,  $(\vec{Q}_X, \vec{s}_Y)$ ,  $(\vec{Q}_Y, \vec{s}_X)$ , and yielded the changes at  $T_F$  shown in solid curves in Fig. 1.

At several fixed temperatures in the  $AF_1$  phase, length changes as a function of measuring field direction were recorded with  $\vec{Q}$  along  $X$  and the field,  $\vec{H} = 22$  kOe, rotating in the  $XY$  plane. These were analyzed on the assumption that for  $\vec{H} \parallel X$  the spins were equally distributed between the  $Y$  and  $Z$  directions and that for  $\vec{H} \parallel Y$  the spins all lay along  $Z$ . Magnetostriction experiments to 100 kOe at 127°K showed that a field of about 40 kOe along  $X$  or  $Y$  and normal to  $\vec{Q}$  is sufficient to saturate the spins into the direction normal to the field and normal to  $\vec{Q}$  and that a field of 22 kOe provides 90% saturation. No correction for lack of saturation has been made to the data for  $\epsilon_s$  or its discontinuity at  $T_F$ . The fact that the data points for  $\epsilon_s$  lie consistently above the dotted

line drawn between the end of the discontinuity at  $T_F$  and zero at  $T_N$  shows that the assumption of equal distribution of spins between the two directions normal to  $\vec{Q}$  is not completely valid and that spin-pinning effects are important.<sup>4</sup>

The results of our measurements are summarized in Fig. 1. The  $\vec{Q}(c_1)$  direction is the long axis just below  $T_N$  and  $c_1/a_1 - 1$  attains a maximum value of  $(4.5 \pm 0.5) \times 10^{-6}$  at about 270°K. However, below about 230°K  $c_1/a_1 - 1$  becomes negative and reaches an extreme value of  $(-20 \pm 2) \times 10^{-6}$  at  $T_F$ . At  $T_F$  the transition from orthorhombic to tetragonal symmetry is accompanied by discontinuities in the lattice parameters. These are  $(c_1 - c_2)/c_1 = (3.5 \pm 0.2) \times 10^{-6}$ ,  $(b_1 - a_2)/b_1 = (-4.7 \pm 0.2) \times 10^{-6}$ , and  $(a_1 - a_2)/a_1 = (-0.2 \pm 0.2) \times 10^{-6}$ . From these values we calculate a volume change at  $T_F$  of  $(v_1 - v_2)/v_1 = -1.4 \times 10^{-6}$  which, using the value of  $dT_F/dP$  reported by Umebayashi et al.,<sup>19</sup> corresponds to a latent heat of  $4.1 \times 10^{-4}$  J/g, in good agreement with the estimate of Street et al.<sup>7</sup> From our measurements we are unable to decide whether the small change  $\Delta a/a$  at  $T_F$  is truly characteristic of the transition or a result of a small number of spins remaining in the measuring direction due to spin-pinning effects. For  $\vec{H} \parallel Z = 50$  kOe we measure  $\Delta a/a = -0.4 \times 10^{-6}$ , whereas for  $\vec{H} \parallel Z = 100$  kOe we measure  $\Delta a/a = -0.2 \times 10^{-6}$ . There is some difference in our measurement of this quantity between the case when  $\vec{Q}$  is along  $X$  and that when  $\vec{Q}$  is along  $Y$ . This suggests further evidence of spin pinning which is also indicated in high-field magnetostriction measurements to be reported elsewhere.

It was found that the depression of  $T_F$  with  $\vec{H} \parallel \vec{Q}$  was quadratic in  $\vec{H}$  and that a field of 100 kOe applied parallel to  $\vec{Q}$  depressed  $T_F$  to about 105°K in good agreement with the work of Street et al.<sup>7</sup> Therefore a temperature of 100°K (below the depressed  $T_F$ ) was chosen for the  $\vec{Q}$ -switching experiment, in which the field was increased from 0 to 100 kOe parallel to  $\vec{Q}$ . Although ideally one would like to perform the  $\vec{Q}$ -switching experiment on a perfect single- $\vec{Q}$  crystal, it appears that increased perfection of the single- $\vec{Q}$  state (achieved by using a higher  $H_c$ ) requires larger fields to achieve any measurable  $\vec{Q}$  switching. For the crystal cooled in  $\vec{H}_c = 50$  kOe along  $Z$  only a very slight irreversible change in length indicative of  $\vec{Q}$  switching was detected. Therefore, the crystal was cooled in  $\vec{H}_c = 35$  kOe along  $Z$ , producing a state largely but not completely single  $\vec{Q}_Z$ . The magnitude of the thermal expansion anomaly at  $T_F$  (in the presence of a field along

$Z$ , so that spins from "wrong  $\bar{Q}$ 's" would not lie in the measuring direction), was taken as a measure of the percentage of the crystal having  $\bar{Q}$  along  $\vec{H}_c$ . This was 80%. Increasing the field from zero isothermally at 100°K produced a continuous irreversible increase in length, beginning at about 45 kOe and reaching a value of  $6.7 \times 10^{-6}$  at 100 kOe, where it was still increasing. The magnitude of the thermal expansion anomaly at  $T_F$  yielded 52% as the percentage of the crystal remaining in  $\bar{Q}_Z$  after  $\bar{Q}$  switching. Normalization of these results to 100% of the crystal gave  $c_2/a_2 - 1 = -24 \times 10^{-6}$ . The entire experiment was repeated for  $\vec{H}_c = 22$  kOe along  $Z$ , yielding 54% before, and 19% after  $\bar{Q}$  switching, and the same value of  $c_2/a_2 - 1$ . These data appear as circled crosses in Fig. 1. The good agreement with the rest of our data suggests that the effect of boundaries between regions of different  $\bar{Q}$  on the external dimensions of a multi- $\bar{Q}$  crystal is small. In any case this  $\bar{Q}$ -switching experiment can be taken as evidence that  $c_2/a_2 - 1$  is negative at 100°K, as it is at  $T_F$ . We are unable to reconcile our observation that  $c_2/a_2 - 1$  is negative down to 100°K with that of Street et al.,<sup>7</sup> who found that a compressive stress applied along  $\bar{Q}$  in the AF<sub>2</sub> phase of a single- $\bar{Q}$  crystal causes the appearance of other  $\bar{Q}$ 's, which at the temperature of our measurements would cause an expansion along the direction of the compressive stress. Perhaps measurements in progress down to liquid-helium temperatures will illuminate this discrepancy.

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