Methods for producing a single wavevector Q state of chromium

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It is shown that the cryomagnetic method prescribed by Golovkin *et al.*, which involves cooling through the spin-flip temperature T_{SF} in a magnetic field along a cube axis, not only fails to produce a single-**Q** state in a chromium single crystal, but tends to destroy the single-**Q** state produced by conventional field-cooling. The failure of the cryomagnetic method is consistent with the anisotropy of the susceptibility in the neighborhood of T_{SF} and other observations.

The conventional method[1,2] of field-cooling to produce a single-wavevector-**Q** state (single-**Q** state) of antiferromagnetic chromium is to cool the single crystal, in a large magnetic field $\mathbf{H}_c$ (the cooling field) parallel to a cube axis, **z** say, through the Néel temperature ($T_N \simeq 311$ K). At some temperature well below T_N, in the transverse spin density wave (TSDW) phase, the field is removed. The resultant state of the Cr sample is single-**Q** to a degree determined by the magnitude of $\mathbf{H}_c$ and the quality of the crystal.

Golovkin *et al.*[3,4] have claimed that their cryomagnetic method (CM) is more effective than conventional field-cooling[1,2] in producing single-**Q** Cr. In this method, which we shall refer to as cryomagnetic-cooling, the field $\mathbf{H}_c$ is applied at room temperature, $T_R \simeq 295$ K, i.e., in the TSDW phase, and the sample is cooled through the spin-flip temperature, $T_{SF} \simeq 123$ K, to liquid nitrogen temperature, $T \simeq 77$ K, i.e., into the longitudinal spin density wave (LSDW) phase. For $\mathbf{H}_c = 2.5$–3.0 T, Golovkin *et al.*[4] found that cryomagnetic-cooling gave a value $I_z/I_x \simeq 25$ for the relative intensities of the satellite neutron diffraction peaks, which is a measure of the relative volumes of the corresponding $\mathbf{Q}_z$ and $\mathbf{Q}_x$ domains, whereas conventional field-cooling for the same sample gave $I_z/I_x \simeq 2$ (the intensity I_y was not measured).

This result is inconsistent with a thermodynamic analysis of the anisotropy of the magnetic susceptibility of Cr in the neighborhood of the spin-flip transition.[5,6] Thus in Fig. 1 we see that the sign of the anisotropy $(\chi_\parallel - \chi_\perp)$ changes at the spin-flip transition. Street *et al.*[6] showed that this change in sign of the anisotropy leads to a reversible depression of the spin-flip temperature T_{SF} proportional to H^2 when a field H is applied along **Q**. They even observed this effect down to $T_{SF}(H) = 95$ K in a field $H = 12.5$ T. They pointed out further that extrapolation of the temperature dependence of $\chi_\parallel$ for the TSDW phase into the LSDW phase, as shown by the dashed line in Figure 1, suggests that the state of lowest free energy can be achieved below $T \simeq 90$ K by a **Q**-flip from **z** to **x** or **y**. Street *et al.*[6] observed this effect with a field, $H = 16$ T, at $T = 77$ K, which irreversibly produced a state having two types of **Q** domains perpendicular to **H**.

A **Q** flip may be induced in lower fields if the sample is not completely one-domain (single-**Q**). Thus Steinitz *et al.*[7] found that a sample field-cooled with $H_c = 5$ T was 80% single-**Q**. This was reduced to 52% by applying $H = 10$ T at a temperature $T = 100$ K along $\mathbf{H}_c$, which had previously been removed at $T \simeq 200$ K. The effect here is presumably due to irreversible growth of the $\mathbf{Q}_x$ and $\mathbf{Q}_y$ domains for $\mathbf{H} \| \hat{\mathbf{z}}$.

Finally, in the course of galvanomagnetic measurements, Arko *et al.*[8] found that at $T = 4$ K, for samples field-cooled with $H_c = 20.5$ T, the single-**Q** state is stable in a transverse field up to $H > 10$ T. If, however, **H** is applied along **Q**, and the electric current flowing through the sample is reversed, an irreversible **Q**-flip occurs, presumably induced by mechanical vibration. If the sample is not completely single-**Q**, a **Q**-flip may occur for $H \simeq 3$–4 T, assisted by vibrating the sample.

It appears therefore not only that cooling through the spin-flip transition with a field $\mathbf{H}_c \| \mathbf{z}$, as in the cryomagnetic method,[4] is unsatisfactory in producing a single-$\mathbf{Q}_z$ state, but that if conventional field-cooling (or application of $\mathbf{H} \| \mathbf{z}$ in the TSDW phase) has provided a predominantly **Q** state, further cryomagnetic-cooling through T_{SF} will tend to cause reversion back to the poly-**Q** state.

We have performed both conventional field-cooling and cryomagnetic-cooling on a high-quality single crystal of Cr and have determined the domain configuration by neutron diffraction. The sample was spark-cut from an arc-zone melted boule and annealed for 72 hours at $T = 1550$ °C. The quality of this crystal is high, with a mosaic spread of only 0.05°. It had previously been measured after field-cooling in a field $\mathbf{H}_c = 12$ T, normally used to prepare the single-**Q** state for ultrasonic velocity measurements.[9,10] As shown in

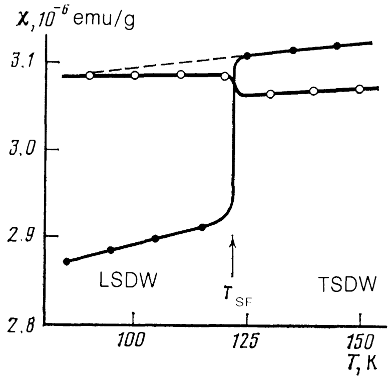

FIG. 1. Anisotropy of the magnetic susceptibility χ of single-**Q** Cr in the neighborhood of the spin-flip transition. ○—Transverse susceptibility $\chi_\perp$ measured along the x axis; ●—longitudinal susceptibility measured along the z axis. $\mathbf{Q} \| \mathbf{z}$.

line 1 of Table I, the sample was then essentially single-**Q**, with $Q = 97\%$ and single-**Q** ratio $R = 65$ (Q and R are defined in footnotes b and c, respectively, of Table I).

A series of measurements was performed, which was designed to check the findings of Golovkin *et al.*,[4] with the results given in Table I. The neutron diffraction data were taken at room temperature, $T_R \approx 295$ K, except for line 4 in which the data for the field-cooled state correspond to a temperature of 148 K in the TSDW phase. The sample was first measured in the nominal poly-**Q** state (line 2 and footnote f in Table I), and this was found to be an accurate description. This result shows that the sample has only small internal strains, with **z** being the preferred axis and $\hat{\mathbf{x}}$, $\hat{\mathbf{y}}$ being roughly equivalent. The z axis was accordingly chosen as the field-cooling and cryomagnetic-cooling direction. In some cases (lines 1, 3, and 4) the satellite intensity I_y (footnote a) was not measured since it could be assumed to be approximately equal to I_x (footnote e).

We see from line 3 that, in this relatively strain-free sample, field-cooling in even as small a field as $H_c = 2.5$ T produces a state with $Q = 63\%$, $R = 3.38$. The sample was restored to the nominal poly-**Q** state by raising its temperature above T_N, and checked experimentally again, as shown in line 2 and footnote f.

The field $H_c = 2.5$ T applied at room temperature produced a state having $Q = 48\%$, $R = 1.80$ (line 4). Thus simply applying the field below and close to the Néel temperature irreversibly increases the fraction of the sample having $\mathbf{Q} \| \mathbf{H}_c$, as found by Golovkin *et al.*[11] On the other hand, we find that cryomagnetic cooling then *reduces* Q to 41%, $R = 1.38$, (line 5), in strong contrast to the result of Golovkin *et al.*[4] who, using the same value of $\mathbf{H}_c$, found that Q *increases* to almost 100%. Thus Golovkin *et al.* [Ref. 4, Fig. 1] give $I = 0.04$, corresponding to $R = I^{-1} = 25$ (footnote c) for cryomagnetic-cooling with $H_c \gtrsim 2.5$ T and starting temperatures $T_H = 303$ K, 295 K, 286 K and 268 K.

Finally, we cooled the sample from above the Néel temperature to below the spin-flip temperature in field, $H_c = 2.5$ T, i.e., we performed a "field-cryomagnetic-cool" (footnote k), which consists of a field-cool followed immediately by a cryomagnetic-cool. The resultant values of Q and R in line 6 are the same as for the nominal poly-**Q** state in line 2, though the distribution between Q_x and Q_y, along the cube axes perpendicular to the field direction, is a little different.

We find therefore that the cryomagnetic-cool effectively destroys the partially single-**Q** state achieved by the field-cool. This result is completely at variance with that of Golovkin *et al.* [Ref. 4, Figure 1, curve 2], who found after field-cryomagnetic-cooling from $T_N = 330$ K (i.e., $T_H > T_N \approx 311$ K) very high values, $Q \approx 93\%$, $R \approx 25$ ($I = R^{-1} \approx 0.04$).

The claim by Golovkin *et al.*[3,4] that the cryomagnetic method is superior to conventional field-cooling is seriously misleading. Thus van Rijn and Alberts[12] followed the prescription of Golovkin *et al.* and, employing a field $H_c = 2.1$ T produced in their Cr sample a state which was quite unsatisfactory for their study of the anisotropy of the elastic moduli.[13] Comparison with the same study performed on our 97% single-**Q** Cr sample[9,10] shows that use of the cryomagnetic method seriously impaired the value of the work of van Rijn and Alberts.

TABLE I. Neutron diffraction analysis of a chromium single crystal at room temperature in zero magnetic field, after various treatments with field H_c (T) along the $\hat{z}$ axis.

	I_x [a] (%)	I_y (%)	$I_z = Q$ [b] (%)	R [c]
1. field-cool[d] in 12 T	1.5	(1.5)[e]	97	65
2. nominal poly-**Q** state[f]	31	33	36	1.12
3. field-cool in 2.5 T	18.5	(18.5)	63	3.38
4. apply 2.5 T at T_R [g]	26	(26)	48	1.80
5. cryomagnetic-cool[h] in 2.5 T	27	32	41	1.38
6. field-cryomagnetic-cooling[k] in 2.5 T	29	35	36	1.13

Notes

[a] I_x, I_y and I_z are the relative intensities of the satellites corresponding to the fractions of the sample having wavevector along the **x**, **y**, and **z** axes, respectively.

[b] Percent single-**Q**: $Q = I_z/(I_x + I_y + I_z)$

[c] Single-**Q** ratio: $R = 2I_z/(I_x + I_y)$; note that $Q = 100\%$ and $R = \infty$ in the ideal single-**Q** state, while $Q = 33\%$ and $R = 1$ in the ideal poly-**Q** state; Golovkin *et al.*[4] define a quantity, $I = I_x/I_z$, such that $I = R^{-1}$ if $I_y = I_x$, as they assume.

[d] Field-cool: cool in $\mathbf{H}_c$ along **z** from some temperature well above the Neel temperature, $T_N \approx 311$ K, to room temperature, $T_R \propto 295$ K.

[e] Values of I_y in parentheses were not measured but were assumed equal to I_x; the justification (and limitations) of this approximation may be understood by inspection of lines 2, 5 and 6.

[f] Ideal poly-**Q** state: $I_x = I_y = I_z = 33\%$; the nominal poly-**Q** state was measured twice after different temperature and field treatments and was found to have the same values of I_x, I_y and I_z within 1%. This provides an estimate of the relative accuracy of our data as being about 3%.

[g] Measured at temperature 148 K.

[h] Cryomagnetic-cool: cool in **H** along **z** from room temperature to some temperature below the spin-flip temperature, $T_{SF} \simeq 123$ K.

[k] "Field-cryomagnetic-cool": cool in **H** along **z** from a temperature above T_N to a temperature below T_{SF}.

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[1] R. A. Montalvo and J. A. Marcus, Phys. Letters **8**, 151 (1964).
[2] A. Arrott, S. A. Werner, and H. Kendrick, Phys. Rev. Letters **14**, 1022 (1965).
[3] V. S. Golovkin, V. N. Bykov, and V. A. Levdik, Pis'ma Zh. Eksp. Teor. Fiz. **14**, 382 (1971) [JETP Lett. **14**, 257 (1971)].
[4] V. S. Golovkin, V. N. Bykov, V. A. Levdik, A. I. Ustinov, and I. P. Barinov, Fiz. Tverd. Tela (Leningrad) **19**, 3439 (1977) [Sov. Phys. Solid State **19**, 2010 (1977)].
[5] A. R. Pepper and R. Street, Proc. Phys. Soc., **87**, 971 (1966).
[6] R. Street, B. C. Munday, B. Window and I. R. Williams, J. Appl. Phys. **39**, 1050 (1968).
[7] M. O. Steinitz, L. H. Schwartz, J. A. Marcus, E. Fawcett, and W. A. Reed, Phys. Rev. Letters, **23**, 979 (1969).
[8] A. J. Arko, J. A. Marcus, and W. A. Reed, Phys. Rev. **176**, 671 (1968).
[9] W. C. Muir, E. Fawcett, and J. M. Perz, Phys. Rev. Letter, 1987 (in press).
[10] W. C. Muir, J. M. Perz, and E. Fawcett, J. Phys. F, 1987 (in press).
[11] V. S. Golovkin, V. N. Bykov, and V. A. Levdik, Fiz. Tverd. Tela (Leningrad) **20**, 1141 (1978) [Sov. Phys. Solid State **20**, 657 (1978)].
[12] H. J. van Rijn, H. L. Alberts, J. Phys. F, 1983, **13**, 1559.
[13] H. L. Alberts, 1987 (private communication).

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