Pressure-Induced "Hardening" of Q-Vector Locking in Chromium

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An irreversible change with pressure of the spin-density wave mismatch parameter δ has been observed in single-Q chromium at 4.2 K. This change, which appears to be analogous to a metallurgical "hardening," can be annealed out by raising the temperature to 80 K. The pressure derivative in the "soft" mode matches that found in high-pressure de Haas–van Alphen studies while the "hard"-mode result is consistent with magnetostriction measurements; so the apparent discrepancy is resolved.

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One of the more striking features of the Fermi surface of single-Q chromium is a chain of ellipsoids whose overlap depends on the incommensurate relationship between the lattice and the Q vector of the spin-density wave (SDW). The majority of de Haas–van Alphen (dHvA) signals derive from breakdown orbits between the hole ellipsoids at N translated over a distance $Q = 2\pi(1 - \delta)/a$. The mismatch parameter δ has a value 0.0484 at 4.2 K and atmospheric pressure,[1] and is independent of temperature up to about 75 K. Its variation with hydrostatic pressure may be determined from the changes in area of the intersection orbits.

The fractional change of $Q' = 1 - \delta$ with pressure derived from magnetostriction studies[2] (equivalent pressure range $\sim 10^{-1}$ bar) is vanishingly small $[(0.01 \pm 0.01) \times 10^{-6}/\text{bar}]$, so that the SDW appears locked to the lattice; whereas that given by comparing dHvA frequencies measured at atmospheric pressure and at several kilobars with use of the solid-helium technique[3,4] is $-(0.50 \pm 0.02) \times 10^{-6}/\text{bar}$, in good agreement with the low-temperature neutron-diffraction value of $-(0.54 \pm 0.05) \times 10^{-6}/\text{bar}$.[5] This large discrepancy has been ascribed to unlocking of the Q vector at some intermediate pressure.[4] In order to investigate this intermediate pressure regime more thoroughly, we have used the fluid-helium dHvA phase-shift technique[6,7] at 4.2 K, at which temperature the pressure range can be extended to $\sim$ 140 bars before solidification occurs.

During the course of these experiments, we observed that the initial application of a given pressure was accompanied by a relatively large, *irreversible* shift of dHvA phase (see Fig. 1 and curve ab in Fig. 2), at a rate corresponding to $d \ln Q'/dP = -(0.55 \pm 0.02) \times 10^{-6}/\text{bar}$, in good agreement with the high-pressure results. Subsequent cycling of the pressure between atmos-

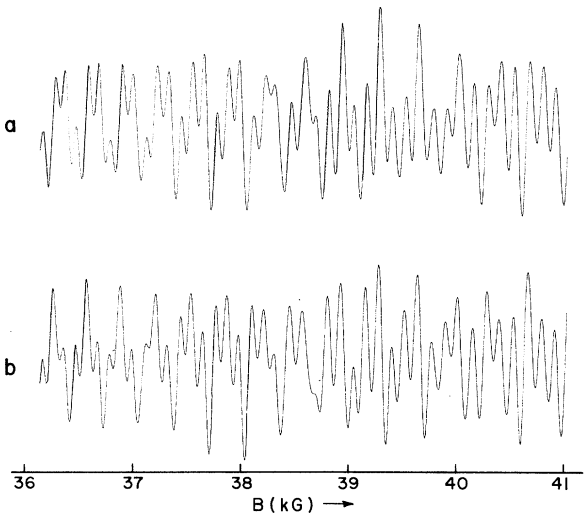

FIG. 1. dHvA traces for Cr between 36 and 41 kG at atmospheric pressure and 4.2 K before (a) and after (b) cycling to 110 bars. The field is perpendicular to the Q vector to within $\frac{1}{2}$ deg. Several frequencies are present, the two dominant ones at 4.3 MG and 12.6 MG both being from breakdown orbits on the ellipsoids. The important feature to note is the change in appearance of the oscillation pattern, seen most clearly near the left edges of the traces. Simple scaling with pressure (orbit areas $\propto 1/a^2$) would give a small, reversible shift of the whole pattern to the right, without changing its appearance. We observe an *irreversible* shift to the *left* (decrease in phase), by a small amount for the high-frequency term and a markedly larger amount for the low-frequency term. This means that both orbits have *decreased* in cross section, the fractional decrease being larger for the smaller orbit.

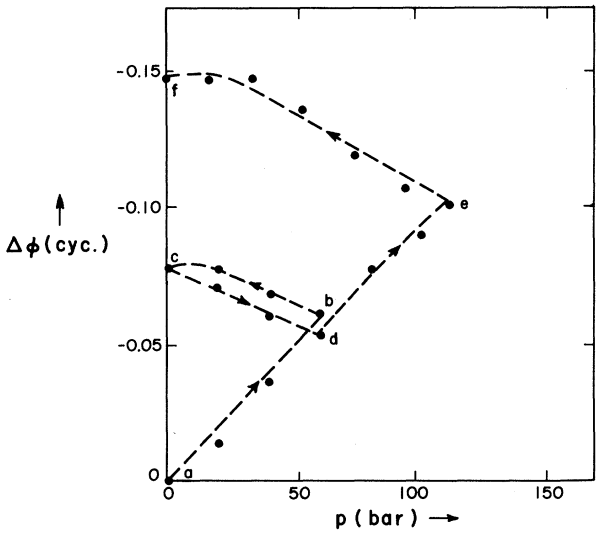

FIG. 2. Absolute dHvA phase vs pressure at 44 kG for the 12.6 MG term. Note that the initial change (curve *ab*) corresponds to a decrease of cross section with pressure; the subsequent changes (segments *bcd*) show an increase of cross section with pressure in the sense and with the approximate magnitude expected for simple scaling, and hence lead to a value of $d\ln Q'/dp$ close to zero.

pheric and the same maximum value showed a *reversible* behavior, with a small hysteresis (segments *bcd*), whose slope on more precise measurement gave a value for $d\ln Q'/dp$ of $(0.005 \pm 0.005)\times 10^{-6}$/bar, confirming the magnetostriction results. However, if the pressure was then increased above the initial maximum, we observed a further irreversible change *at the same rate as before* (curve *de*), followed again by the reversible behavior on cycling (curve *ef*).

This suggested that we were observing a progressive locking of the Q vector into some strain-dependent final state, analogous to a metallurgical hardening process, and that it might be possible to "anneal out" this strained condition by a suitable increase of temperature. Accordingly, we raised the sample holder out of the liquid helium bath, allowing it to reach a temperature of, perhaps, 50–100 K, estimated by monitoring the dHvA modulation coil resistance. On replacing the probe we found that the specimen had reverted to its original, unpressurized condition. Since this procedure also involved removing the sample from the ~40-kG magnetic field, we checked that removal of the field alone allowed no detectable relaxation to occur. In order to eliminate the possibility that the observed effects were due to a mechanical shift of the sample assembly under pressure, we replaced the Cr sample with one of Sn,[8] and confirmed that there was no deviation from a linear relationship between dHvA phase and pressure over the full pressure range. A further measurement, suggested by the author of the model discussed below,[9] involved cooling the sample to 4.2 K with the pressure already partially applied. We observed the "hard" mode when the pressure was reduced, and the "soft" mode when it was first increased from the initial value.

Having shown that thermal relaxation of the "hardened" state was possible, we then attempted to determine the "softening" temperature range more precisely. Since the pickup coil of the dHvA probe was closely coupled to the pressure capsule and fairly well isolated from the helium bath it was possible to use it both as heater and thermometer. Satisfactory annealing was achieved by this technique, as little as 10 sec at a given voltage/current combination being sufficient to achieve a relaxation characteristic of that temperature. The degree of annealing remained unchanged during further heating to the same temperature. Relaxation, as observed from the dHvA waveform at ~36 kG, began a little below 30 K and was essentially complete by 80 K.

It should be noted that the solid-helium pressure technique, used for the high-pressure dHvA measurements,[3,4] requires that the system be warmed up to some 80 K before changing the pressure, so the measurements will always give results corresponding to a "virgin" sample. On the other hand, magnetostriction, which involves continuous cyclic strain, will always be measuring a sample that is hardened to the limits of the magnetostriction. The existence of a "hardening" phenomenon thus provides a simple explanation of the discrepancy between the results obtained by the two techniques.

It has been suggested[10] that impurities and dislocations may be responsible for "pinning" the Q vector. While this might well cause friction between the spin-density wave or its associated charge-density wave and the lattice, it is difficult to see how it could allow for the observed easy slippage *in one direction only*—analogous to a ratchet mechanism. Furthermore, any annealing associated with dislocations would be most unlikely to occur at a temperature as low as 50 K. We discuss here an alternative idea[9] which appears to have the required characteristics.

According to this model, domain-wall-like phase changes occur in the periodicity of the

crystal lattice relative to the SDW phase, without changing the phase or Q vector of the more rigid SDW. This mechanism allows the combined crystal lattice and itinerant electron SDW energy to be lowered. The free energy of these domain walls is negative, so they can be created readily but require thermal energy for their destruction. An increase of δ with increase in pressure is accompanied by generation of additional domain walls, because N_{DW}, the number of domain walls per centimeter in the direction parallel to Q, is almost exactly proportional to δ. If the temperature is sufficiently low, the extra domain walls will be retained when the pressure is decreased, which means that no decrease in N_{DW} or in δ can occur. Subsequent cycling of pressure will cause no additional change unless the initial maximum pressure is exceeded, and relaxation will not occur until sufficient thermal energy is available for the new domain walls to be destroyed.

In order to avoid significant reduction of the dHvA signal, it is essential that phase coherence of the lattice be maintained across the domain walls, and that electron scattering by the domain walls be small. Provided that these conditions are met, this mechanism appears to satisfy the requirement of a "thermodynamic ratchet" to explain our experimental observations.

A number of additional experiments based on the proposed model have been suggested to us,[9] in particular to observe the effect of changing the specimen dimensions parallel to Q, and to study dilute alloys of, e.g., manganese in chromium, where the zero-pressure mismatch parameter δ is different from that in the pure metal.

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Domains in the Spin-Density-Wave Phases of Chromium

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Lattice-distortion domain walls in the spin-density-wave phases of Cr are thin, have negative free energy, and provide an explanation for the irreversibility of increases of δ in $Q = (2\pi/a)(1 \pm \delta)$ with increases in pressure at $T \ll 50$ K, observed in de Haas–van Alphen experiments of Ruesink, Perz, and Templeton. Domain walls of the spin-density wave itself have positive free energies, have thickness $\gtrsim 1000$ Å, and cannot be used to explain these experimental results.

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Ruesink, Perz, and Templeton have observed a new and unexpected behavior of the Q vector in chromium under pressure.[1] When a Cr crystal is cooled to 4.2 K under atmospheric pressure and is subsequently subjected to larger pressures, while δ in $Q = (2\pi/a)(1 \pm \delta)$ of the spin-density wave (SDW) (where a is the lattice constant) is measured via de Haas–van Alphen techniques, $\delta(P)$ increases from $\delta(0) \approx 0.05$ at the rate $d \ln\delta/dP \approx 9.5 \times 10^{-6}$ bar^{-1} as long as the pressure is monotonically increased. If the increase is stopped at any P_{max} and the pressure then decreased to any low-