The charge density wave instability in uranium

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Uranium is the only element known so far to exhibit a charge-density wave instability. The structure below the transition (T_0 ~ 43 K) is complex consisting of small displacements of the atoms away from their α-phase equilibrium structure position. It is usually assumed that this transition involves a soft-phonon condensing at (or near) the charge-density wave position in the Brillouin zone. New high-resolution neutron scattering experiments at low temperature show that the principal softening occurs *exactly* at the charge-density wave positions, as expected for a pure Fermi-surface instability. This result is in disagreement with a model of Yamada (1993), but in agreement with a recent theory which suggests that the charge-density wavevector components are related to the Fermi-surface nesting topology.

The low-temperature phase of uranium metal (the alpha phase is stable below 935 K) has been known for many years to exhibit a charge-density wave (CDW) below T_0 ~ 43 K, at which temperature many physical properties show unusual anomalies [1]. Discovered using Laue diffraction almost twenty years ago [2] at the ILL, α-U is the only element, so far, to exhibit such a transition. The pattern of atomic displacements that appears just below T_0 is *incommensurate* in all three crystallographic directions in the orthorhombic unit-cell. More precisely, $\mathbf{q}_{CDW} = [q_x\mathbf{a}^*, q_y\mathbf{b}^*, q_z\mathbf{c}^*]$, where $\mathbf{a}^*$, $\mathbf{b}^*$, $\mathbf{c}^*$, are the reciprocal lattice vectors of the orthorhombic parent α structure, and at T_0

$$\mathbf{q}_{CDW} = \pm 0.490\,\mathbf{a}^* \pm 0.131\,\mathbf{b}^* \pm 0.225\,\mathbf{c}^*. \quad (1)$$

At temperatures below T_0 all three q-components become (with first-order transitions) *commensurate*. These lower temperature transitions can be understood on the basis of attempting to minimise the strain energy in the crystal, but the unusual initial values remain difficult to understand. An additional mystery was that around T_0 (and remaining to the lowest temperatures) a weak peak was observed at the position $\mathbf{q}' = [q'_x, 0, 0]$, where $q'_x \sim 1/2$.

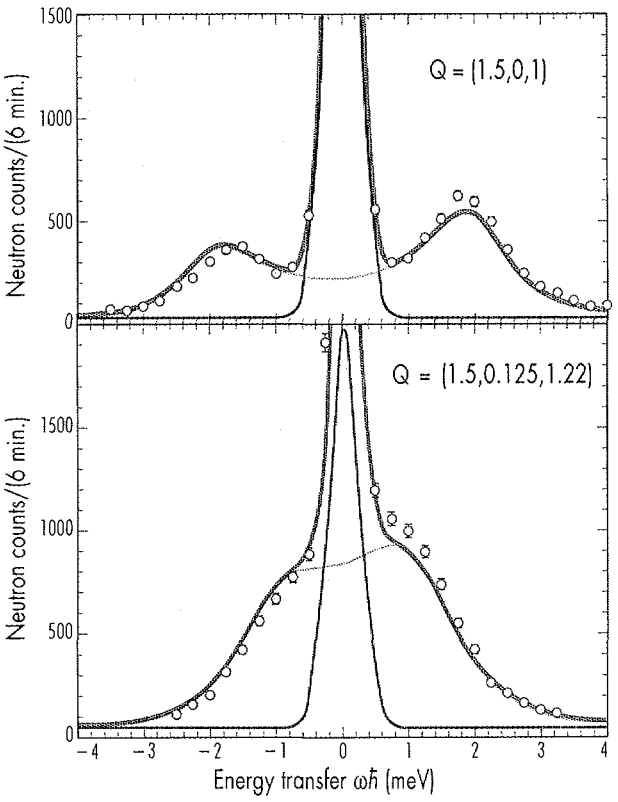

Figure 1: *Constant-Q spectra of α-U from IN14 for two momentum transfers with the same value of q_x = 0.5, but the top frame (Q_1) has $q_y = q_z$ = 0, whereas in the lower frame at Q_2 these have non-zero values (T = 55 K, K_f = 2.2 Å^{-1}). The green lines are fits to the data, with the phonon and elastic contributions shown as orange and red lines, respectively.*

In 1993, Yamada [3] advanced a theory that the additional peaks at $\mathbf{q}'$ were a consequence of strain in boundary regions between the different domains of the main charge-density wave distortion, occurring at $\mathbf{q}_{CDW}$. While Yamada's theory is successful in explaining this particular feature of the low-temperature charge-density wave state, it also postulates that the main driving force leading to the transition, via the softening of the Σ_4 phonon, depends *only* on interactions in the [100] (q_x) direction. This implies a dispersion surface of the Σ_4 phonon that is parabolic in q_x and has no dependence in the (q_y, q_z) plane. The fact that $\mathbf{q}_{CDW}$ has finite components in q_y and q_z is viewed as yet another consequence of the coupling of the charge-density wave displacements with the strain.

The motivation for our experiments was to test these ideas in α-U, and to learn more about the soft-mode dispersion in the three orthogonal directions around $\mathbf{q}_{CDW}$. The experiments used both the IN14 and IN8 three-axis spectrometers to follow the phonon frequencies from room to low temperature. Here, we will concentrate on the results from IN14. Figure 1 shows phonon groups for two values of momentum transfer with the same q_x. It is clear that the phonon frequency is higher in the top frame than it is in the lower frame. Since q_x = 0.5 in both, and Yamada's theory [3] conjectures that the frequency should depend on q_x only, this part of the theory is incorrect.

A more refined analysis was performed assuming that a real minimum exists in the dispersion surface $\mathbf{q}_{min}$, and that the phonon frequency is quadratic between $\mathbf{q}_{min}$ and any measu-

red point **q**. This analysis, involving a 4-dimensional convolution with the instrumental resolution function, was performed on a total set of 11 energy scans with different **q**'s at 55 K. The resulting dispersion surface is shown in Fig. 2. This figure shows that the dispersion surface depends on both q_y and q_z, at variance with the theory of Yamada. Furthermore, the value of $\mathbf{q}_{min}$ is given by

$$\mathbf{q}_{min} = \pm 0.485(5)\,\mathbf{a}^* \pm 0.13(1)\,\mathbf{b}^* \pm 0.21(1)\,\mathbf{c}^*. \quad (2)$$

Perhaps surprisingly, $\mathbf{q}_{min}$ (measured here above T_0) is indistinguishable from $\mathbf{q}_{CDW}$ (see Eq. (1)) as determined from diffraction experiments just below T_0. This implies that the phonon is already sensing the instability before it occurs. Such an interaction is almost certainly a signature of a Fermi-surface instability. Recently, Fast et al. [4] have developed a theory incorporating the itinerant nature of the 5f electrons and shown that the resulting Fermi surface does have some unusual features. Our data can be understood as a direct measure of these Fermi-surface nesting effects sensed through the (electron-phonon driven) phonon softening (Kohn anomaly).

Finally, assuming that $\mathbf{q}_{min}$ does not change appreciably as a function of temperature, which is important to verify in the future, we can obtain the temperature variation of the frequency minimum at $\mathbf{q}_{min}$. This is plotted, together with values measured at two other momentum transfers, in Fig. 3. The soft frequency follows a $(T - T_0)^{0.5}$ law with an intercept at T_0 [5], as expected in the classical Landau theory.

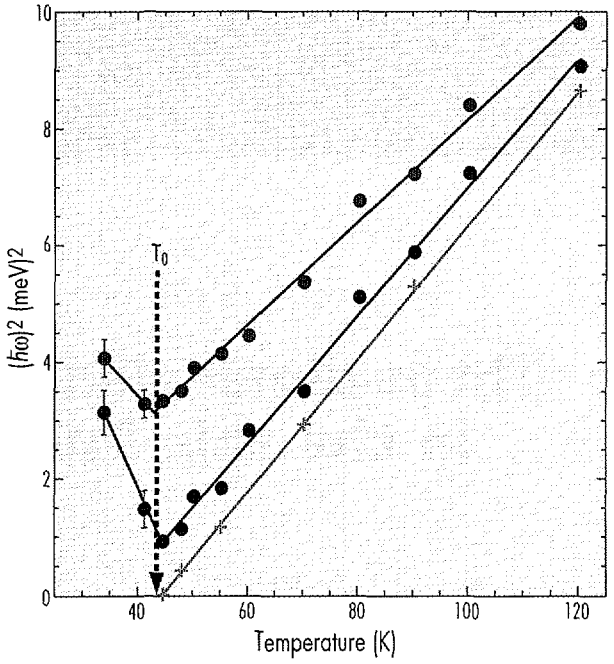

Figure 3: *The square of the soft-mode oscillator frequency plotted as a function of T at positions near momentum transfers Q_1 = [1.5, 0, 1] (red circles and as shown in upper frame in Fig. 1) and Q_2 = [1.5, 0.125, 1.22] (purple circles and as shown at T = 55 K in lower frame of Fig. 1). The crosses correspond to the values at q_{min} as extracted from the fits described in the text.*

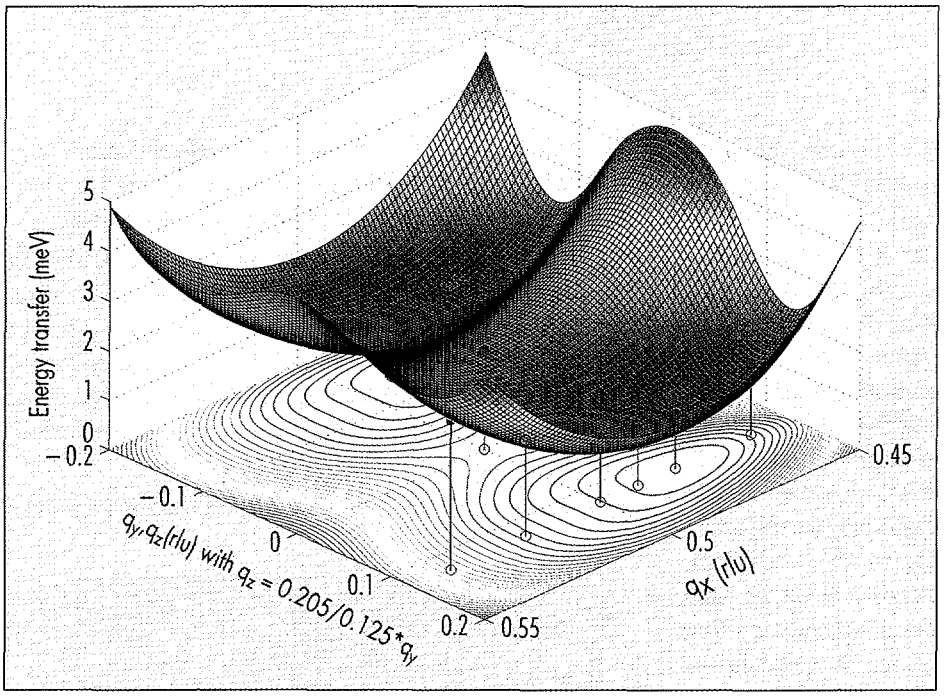

Figure 2: *A three-dimensional view of the dispersion surface projecting along an oblique direction in the (q_y, q_z) plane in order to show the minimum position – see definition of q_{min} in text. The red circles show the experimentally measured phonon frequencies at T = 55 K.*

References

■ [1] For a review of work up to 1993 see G. H. Lander, E. S. Fisher, and S. D. Bader, Adv. Physics 43 (1994) 1

■ [2] J. C. Marmeggi and A. Delapalme, Physica B 120 (1980) 309 ■ [3] Y. Yamada, Phys. Rev. B 47 (1993) 5614

■ [4] L. Fast, O. Eriksson, B. Johansson, J. M. Wills, G. Straub, H. Roeder, and L. Nordström, Phys. Rev. Lett., 81 (1998) 2978

■ [5] J. C. Marmeggi, R. Currat, A. Bouvet, and G. H. Lander, Proc. Int. Conf. on Phonons July 98 Lancaster, UK, Physica B (263-264) 1 (1999) 624.