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Phonon anomalies in α-uranium

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Abstract

The temperature dependence of the phonon spectra of α-uranium has recently been measured by inelastic neutron scattering and inelastic X-ray scattering. Although there is little evidence of any anharmonicity, the phonon shows some softening in the optic modes at the zone boundary. Furthermore, an extra mode forms at high temperatures, which is incompatible with a structure composed of a monoclinic Bravais lattice with a two-atom basis. We investigate the effect that the f electron–phonon interaction has on the phonon spectrum and its role on the possible formation of a breathing mode.

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Keywords: α-uranium; Phonon anomaly; Breathing mode

1. Introduction

The properties of uranium and cerium compounds often exhibit similarities. Both sets of compounds span the border of magnetism and non-magnetism, and also both may exhibit heavy fermion phases. The anomalous magnetic properties of these compounds are connected to fluctuations of the f occupation number which also produce lattice anomalies, such as those found in the $\alpha \rightarrow \gamma$ transition in elemental cerium [1,2]. Anomalies have been found in the phonon spectra of α-uranium measured by inelastic neutron scattering [3,4] in the temperature range of 50–1213 K. Although there is scant evidence of anharmonicity, the phonon spectrum was observed to soften with increasing temperature. The softening is primarily in the optic modes near the Y point of the Brillouin zone. Furthermore, an extra mode was found [5] to evolve above 450 K, which is incompatible with the harmonic phonons of a structure with a monoclinic unit cell with a two-atom basis. Since no structural transitions have been observed in the temperature range 298–573 K, and since the charge density waves in uranium [6] occur at temperatures below 60 K, it seems unlikely that the phonon anomalies are due to these transitions. Therefore, we shall examine the effects of strong electron–phonon coupling, and investigate the existence of optical breathing modes with low symmetry.

2. Model

We model the 5f electronic states in α-uranium as being localized, but are coupled to the conduction band by an on-site hybridization process. The phonon spectrum, excluding the f electron–phonon coupling, is shown in Fig. 1, for the $[0,1,0]$ and $[0,0,1]$ directions. A large Kohn-anomaly has been observed in the longitudinal optic mode [7] at $(0.5,0,0)$ and was found to soften as the temperature is reduced towards the charge density wave instability [6]. The presence of the Kohn anomaly signifies the importance of electron–phonon interactions. We introduce an f electron–phonon interaction $\lambda_{q,\alpha}$ through the effect of the size of the uranium atoms [1]. We consider the limit in which the f states are just above the Fermi-energy. The f-electron phonon coupling is treated by performing a unitary transformation

$$\hat{U} = \exp\left[-\sum_{j,\underline{q},\alpha} \frac{\lambda_{\underline{q},\alpha}}{\hbar\omega_{\underline{q},\alpha}} (a^{\dagger}_{\underline{q},\alpha} - a_{-\underline{q},\alpha}) f^{\dagger}_{j} f_{j} \exp[i\,\underline{q}\,.\underline{R}_{j}]\right], \qquad (1)$$

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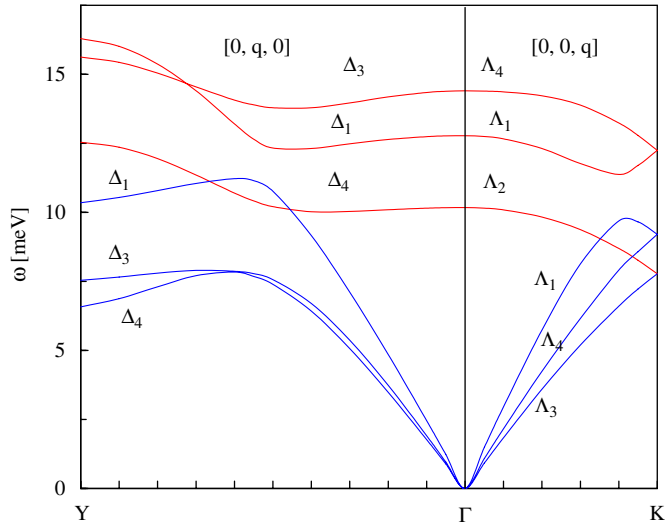

Fig. 1. The phonon dispersion relations for the α-uranium structure, excluding the coupling to the f electrons.

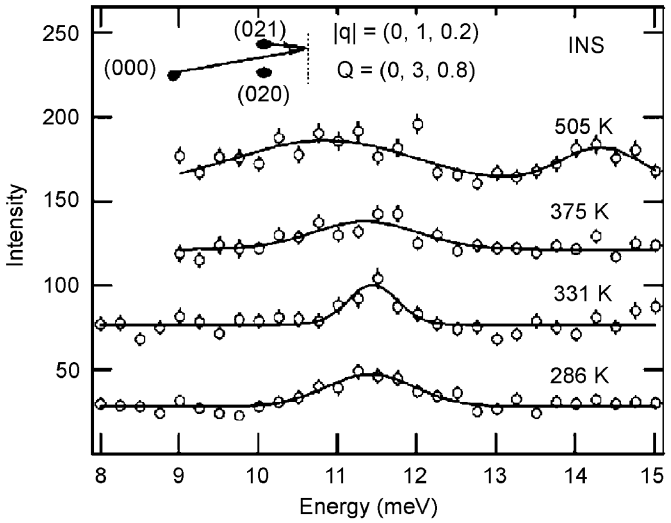

Fig. 2. The experimental data of Manley et al. Ref. [3]. showing the evolution of a new mode at $(0, 1, q)$ near the Y-point.of the Brillouin Zone boundary.

where $a^{\dagger}_{\underline{q},\alpha}$ and $a_{\underline{q},\alpha}$ are, respectively, the creation and annihilation operators for a phonon of frequency $\omega_{\underline{q},\alpha}$ while $f^{\dagger}_j$ and f_j are the creation and annihilation operators for f electrons at site j. The canonical transformation produces a Friedel interaction between the occupancies of the ions at site i and j given by

$$\hat{H}_{\rm int} = -\sum_{i,j,\underline{q},\alpha}\left(\frac{\lambda^2_{\underline{q},\alpha}}{\hbar\omega_{\underline{q},\alpha}}\right)\exp[i\underline{q}.\underline{R}_{ij}]f^{\dagger}_i f_i f^{\dagger}_j f_j. \tag{2}$$

This interaction has the tendency of promoting charge ordering. The canonical transform also has the result of dynamically renormalizing the f electron hybridization interaction matrix elements via

$$\sum_{\underline{k},j} V(\underline{k}) f^{\dagger}_j d_{\underline{k}} \exp\left[\sum_{j,\underline{q},\alpha}\frac{\lambda_{\underline{q},\alpha}}{\hbar\omega_{\underline{q},\alpha}}(a^{\dagger}_{\underline{q},\alpha} - a_{-\underline{q},\alpha})\exp[i\,\underline{q}\,.\underline{R}_j]\right]. \tag{3}$$

The thermal average of the matrix element exhibits a polaronic reduction

$$V_{\rm eff} = V\exp\left[-\frac{1}{2}\sum_{\underline{q},\alpha}\left(\frac{\lambda_{\underline{q},\alpha}}{\hbar\omega_{\underline{q},\alpha}}\right)^2(1+2N_{\underline{q},\alpha})\right], \tag{4}$$

where $N_{\underline{q},\alpha}$ is the Bose–Einstein distribution function. The dynamical hybridization matrix element and its Hermitean conjugate are used to calculating the phonon self-energy $\Pi_{\alpha}(\underline{q},\omega)$ to nominally second-order in the dynamical hybridization matrix element. It should be noted that phonon propagation between pairs of vertices partially removes the polaronic reduction. However, the quasi-particle peak remains fully renormalized. This polarization part describes a process in which a phonon scatter an f electron into the conduction band and vice versa, and as a result its imaginary part has a double peaked structure which reflects the hybridized electronic density of states. The phonon frequencies are determined as the solutions of the equation

$$\omega^2 = \omega^2_{\underline{q},\alpha} + 2\omega_{\underline{q},\alpha}\Pi_{\alpha}(\underline{q},\omega) \tag{5}$$

in which the imaginary part is set to zero. Since the real and imaginary parts of $\Pi(q,\omega)$ are related through a Hilbert transform, we find that for a limited range of the hybridization, it is possible to have two solutions to the real part of the above equation. These two solutions describe excitations of mixed electron and phonon characters. This mixing is expected to be found at energies where the hybridized f electron density of states shows structure very close to the Fermi-energy, and so should primarily affect the high-energy phonon modes. The new high-energy solution may be characterized as a breathing mode which involves the phonons coupling coherently with f–d charge fluctuations. The width of the phonon lines are proportional to the imaginary part of the polarization part at the respective frequencies, and results in the solution with lower frequency experiencing a larger broadening than the higher-frequency solution. Furthermore, on decreasing the hybridization, the two peaks coalesce. We tentatively identify these two solutions with the formation of the extra mode found by Manley et al. [5] depicted in Fig. 2. Since the calculated phonon dispersion relations shown in Fig. 1 are highest near the Y point, it is expected that these modes should show the most coupling. This is in agreement with the experimental findings of Manley et al. from which it appears that only the highest-energy optic modes are softened.

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References

[1] D.S. Sherrington, P.S. Riseborough, J. Phys. Paris 37 (1976) C4 p255.
[2] S. Basu, P.S. Riseborough, Physica B 378–380 (2006) 686.
[3] A.C. Lawson, B. Martinez, J.A. Roberts, B.I. Bennett, J.W. Richardson Jr., Philos. Mag. B 80 (2000) 53.
[4] M.E. Manley, B. Fultz, R.J. McQueeney, C.M. Brown, W.L. Hults, J.L. Smith, D.J. Thoma, R. Osborn, J.L. Robertson, Phys. Rev. Lett. 86 (2001) 3076.
[5] M.E. Manley, M. Yethiraj, H. Sinn, H.M. Volz, A. Alatas, J.C. Lashley, W.L. Hults, G.H. Lander, J.L. Smith, Phys. Rev. Lett. 96 (2006) 125501.
[6] H.G. Smith, N. Wakabayashi, W.P. Crummett, R.M. Nicklow, G.H. Lander, E.S. Fisher, Phys. Rev. Lett. 44 (1980) 1612.
[7] W.P. Crummett, H.G. Smith, R.M. Nicklow, N. Wakabayashi, Phys. Rev. B 19 (1979) 6028.