Understanding the volume collapse transition in $Ce_{0.9}Th_{0.1}$

Strange phonon behavior, magnetic excitations and an interesting competition

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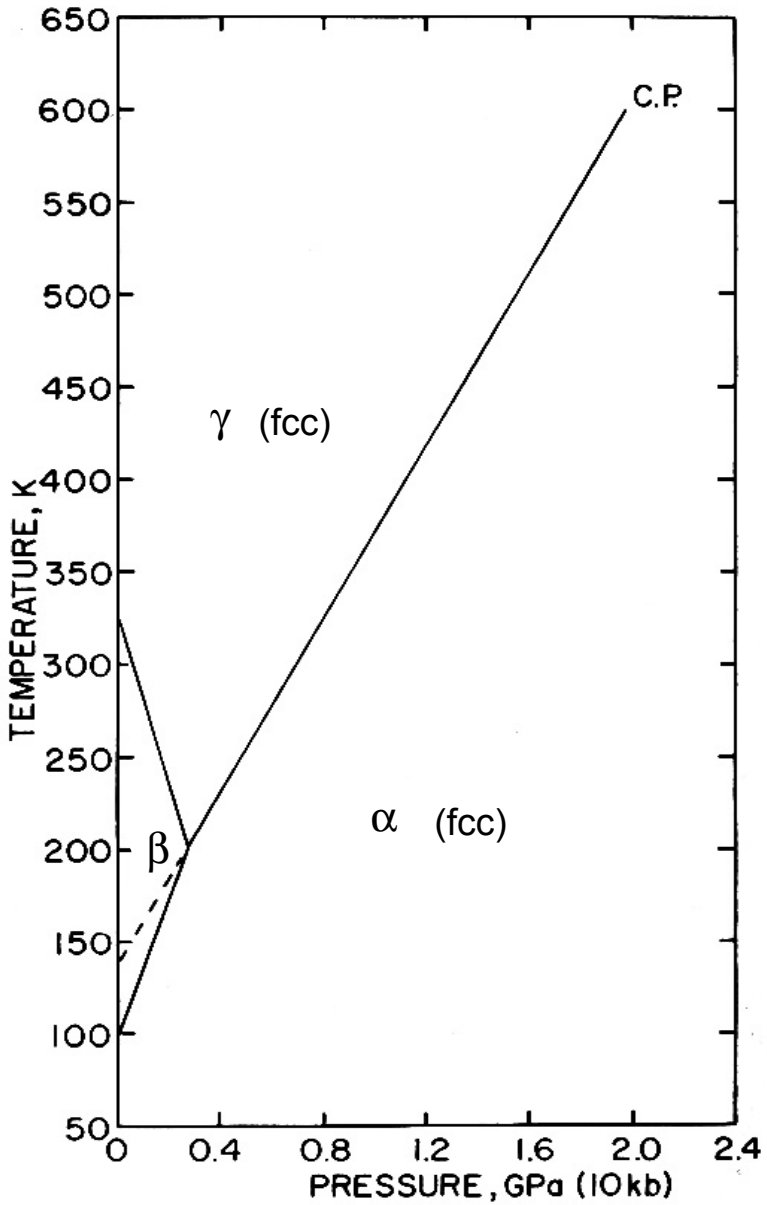
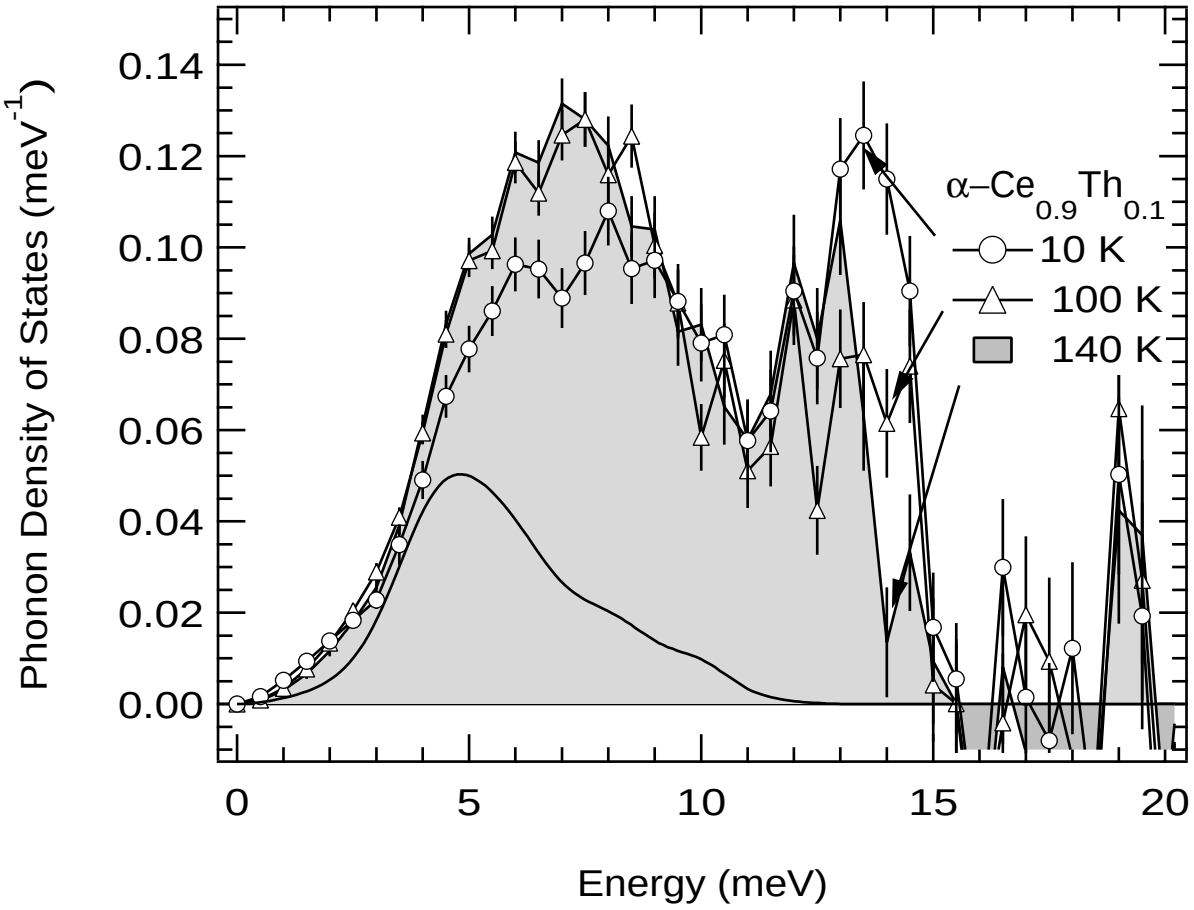
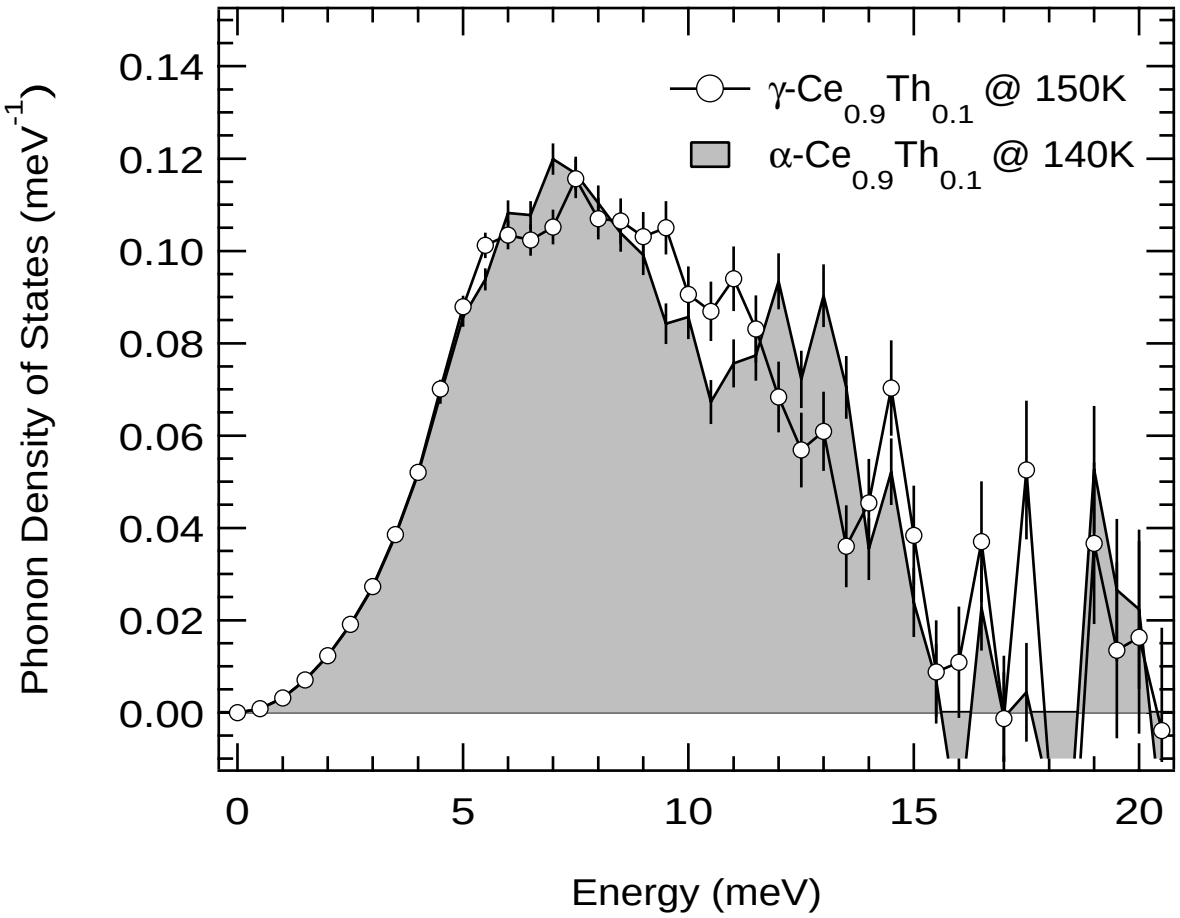
Cerium temperature-pressure phase diagram



Phonon density of states of the collapsed α-phase

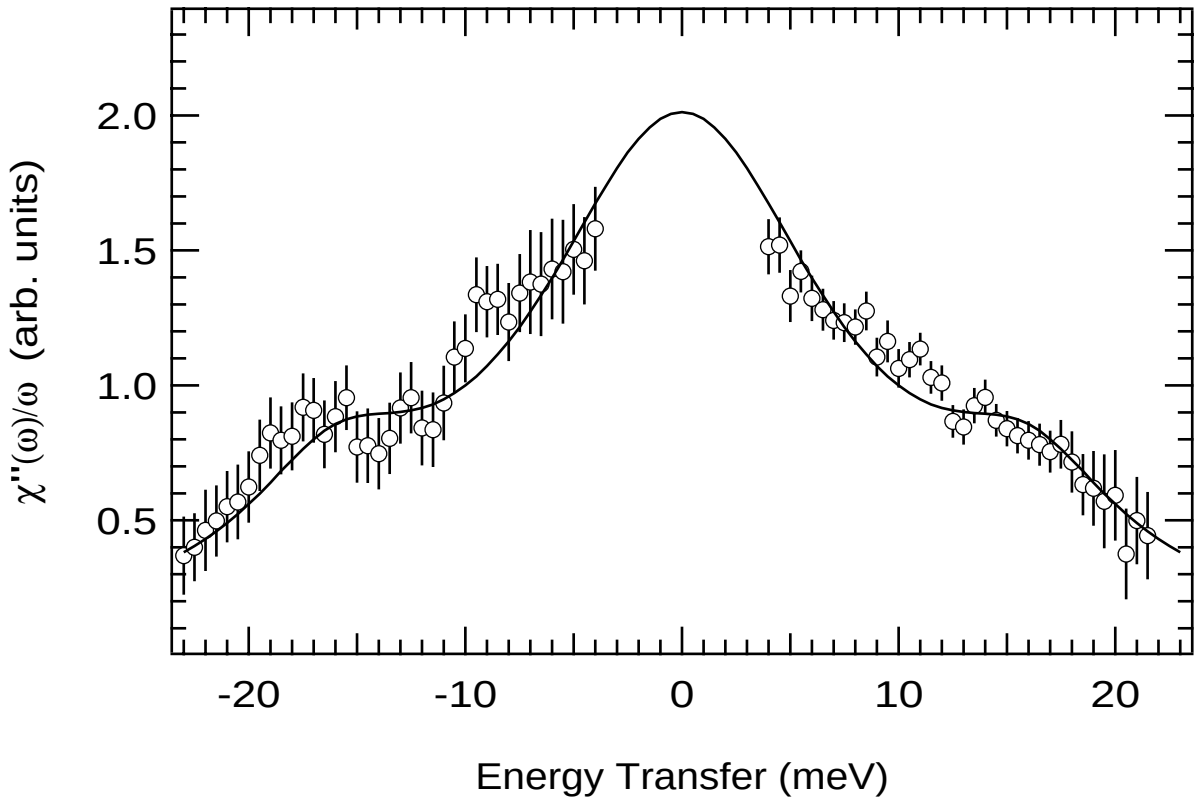


Phonon density of states before and after a 17% volume change



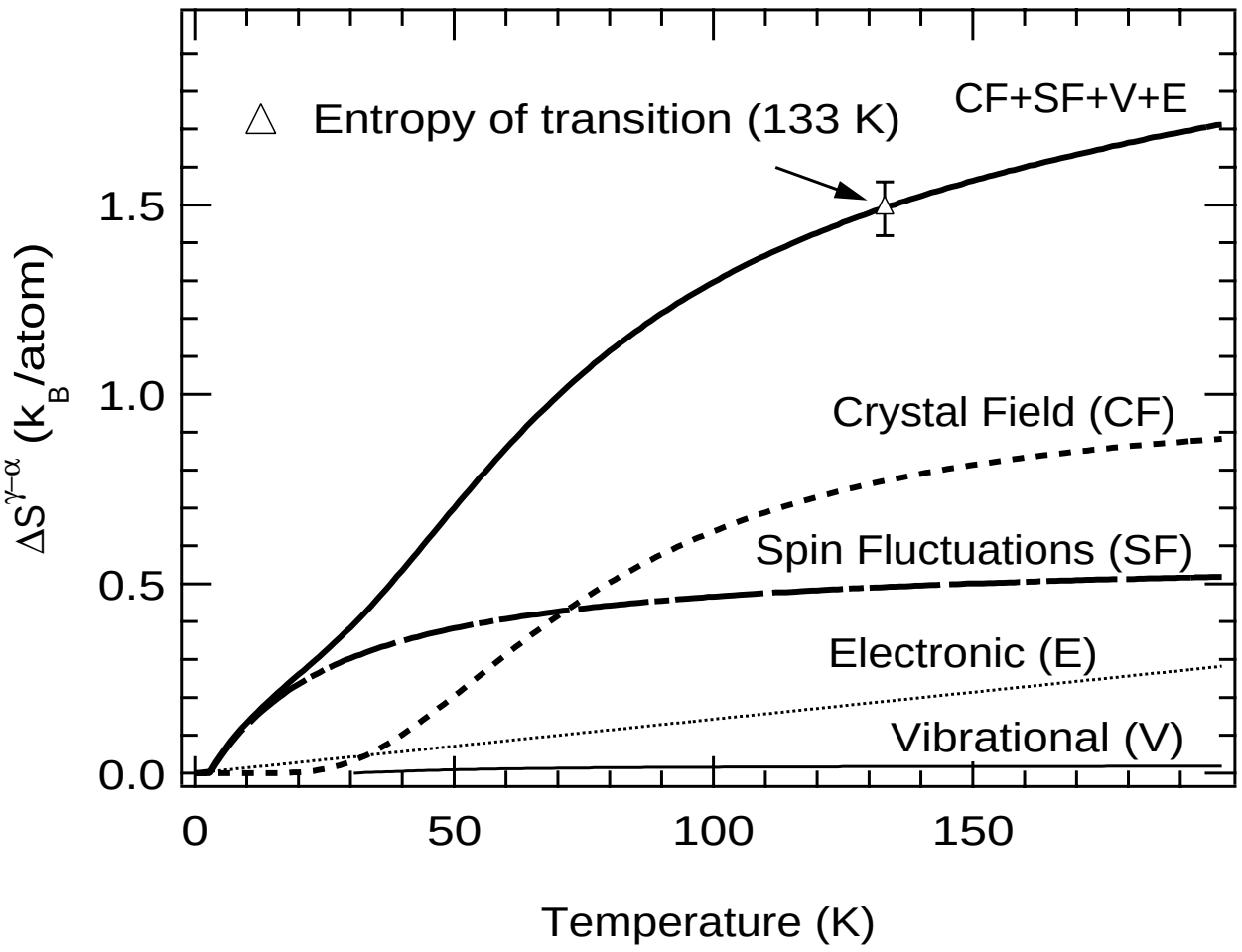
Anharmonic expectation: $\frac{\delta\omega}{\omega_0} = -\gamma\frac{\delta V}{V_0} \cong 0.34$

Dynamic magnetic susceptibility of the expanded γ-phase



Quasielastic width ~ 170 meV in α-phase [Murani et al. PRB **48** (1993)]

Transition entropy can be explained without phonons



Anharmonic versus Kondo-like spin fluctuation effects on elastic properties

Grüneisen expression for anharmonic effect: $B_{anh} \cong B_0 (V_0 / V)^{\gamma}$ ($\gamma \approx 2$, B_0 = 28 GPa)

Spin fluctuation contribution:

Free energy from spin fluctuations

$$F_{SF}(V,T) = E_{SF} - TS_{SF} = \int_0^T C_{SF}(V,T')dT' - T\int_0^T \frac{C_{SF}(V,T')}{T'}dT'$$

$$\Rightarrow B_{SF}(T) = V\frac{\partial^2 F_{SF}}{\partial V^2} = V\int_0^T \frac{\partial^2 C_{SF}(V,T')}{\partial V^2}(1 - T/T')dT'$$

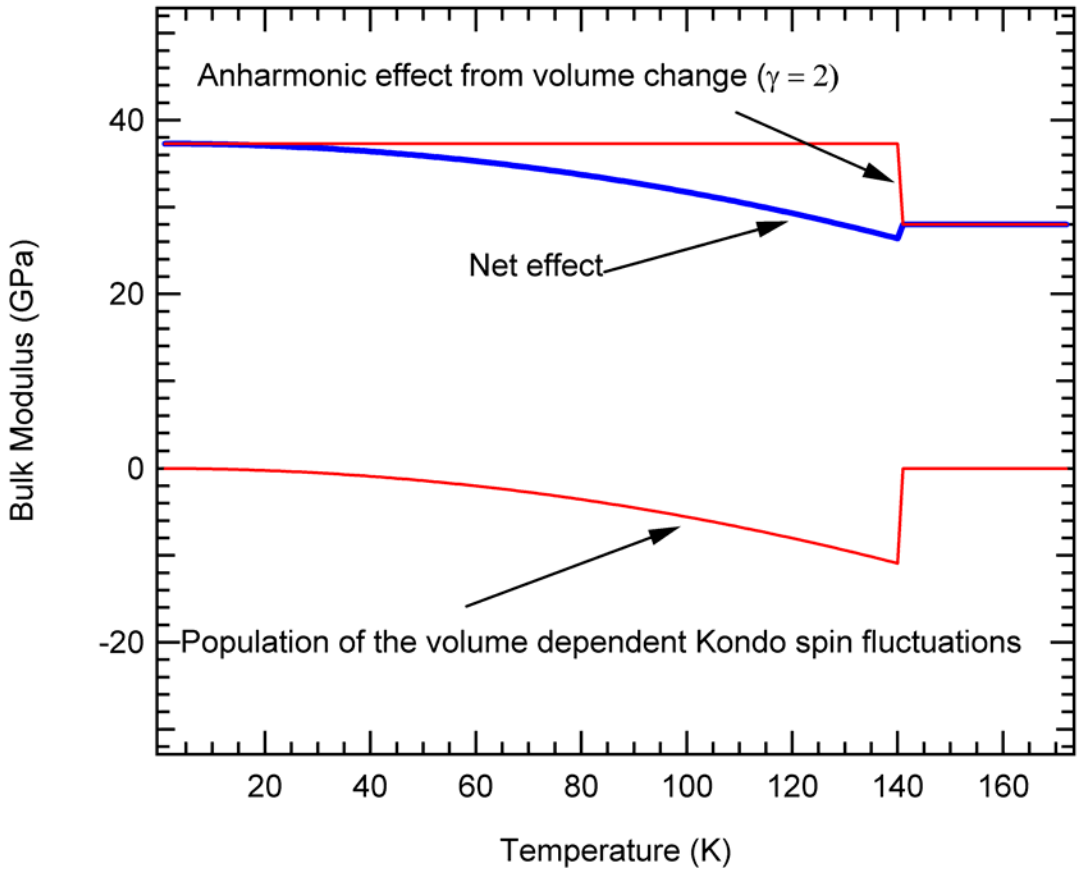
Using an exact expression for C_{SF} [Rajan PRL **51** (1983)]

$$B_{SF}(T \to 0) \cong -(1.65/\pi)(N-1)k_B^2 V \frac{\partial^2}{\partial V^2}\left(1/\Gamma(V)\right)T^2$$

To get numbers we assume

$$\Gamma(l) = 170(1 + \beta(l - l_0)/l_0)^{-n} \qquad (l^3 = V)$$

Anharmonic and Kondo (electron correlation) effects cancel at the transition



Result: The $\alpha \rightarrow \gamma$ transition in $Ce_{0.9}Th_{0.1}$ is a massive volume collapse driven purely by electronic degrees of freedom!

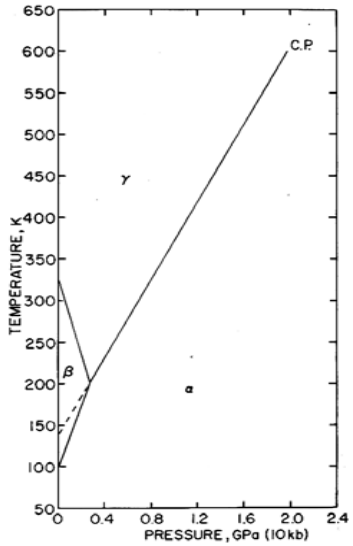
Conceptual picture for cerium

Conduction electrons

Localized f-electrons

Ground state spin fluctuations (SF)

kT



Strange phonon behavior

SF energy

SF & CF entropy gained by volume expansion drives transition

$$G = E + PV - TS$$

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