Supplementary Information

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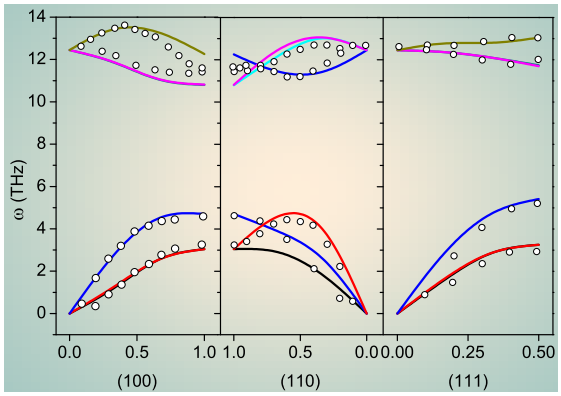
Phonons and Electron-Phonon Interactions

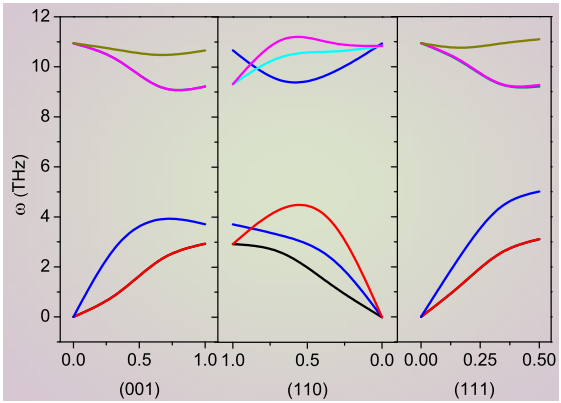
For the calculation of phonon spectra we used density functional linear response approach implemented using LMTO basis [1, 3], which has successfully produced the lattice dynamics of solids [4] including actinides materials such as plutonium [9] and UO_2 [7]. The spin–orbit coupling effect is included in this calculation. A $\mathbf{q}$–grid of $6\times6\times6$ is used to compute phonon frequencies, which generates 36 irreducible q points in the Brillouin zone. The phonon dispersion of UN along 3 high–symmetry directions is plotted in Fig. 1(a) together with experimental data measured by neutron scattering [10]. As shown in Fig. 1(b), UC carries similar phonon dispersions but slightly lower phonon energies. Despite apparent presence of correlation effects, excellent agreement is achieved with the local density approximation (LDA). Similar success of LDA in studying lattice dynamics of strongly correlated metallic systems have been reported earlier, for example in Palladium [4], high temperature superconducting cuprates [5], and recently iron pnictides [6].

Calculations of electron–phonon interactions and transport properties require, on the other hand, quasiparticle description of the one–electron spectra when evaluating Eliashberg and transport spectral functions by integrating over the Fermi surfaces [4]. As a result, due to large mass enhancement, the straightforward LDA procedure can produce wrong electron–phonon resistivities which was indeed found in our calculation for UC where $\rho(T)_{EPI}$ was overestimated by a factor of 3 compared to experiment. This is despite of simple arguments that would suggest that any multiplicative effects on the electron mass renormalization should cancel out in the resistivity, because it enters both the scattering rate τ that appears in the denominator, and the electronic mass that appears in the numerator of the expression for $\rho(T)_{EPI}$, which is evident from a simple Drude formula for $\rho = m/(ne^2\tau)$. However, in general, this does not apply to multi–band systems where only correlated f–electron wave functions are primarily affected by strong Coulomb interactions.

In order to evaluate the electron–phonon scattering in the presence of correlations we develop a method that accounts for the effects from quasiparticle mass renormalizations and spectral weight transfer by utilizing interacting Green functions. We have previously shown [8]



(a)



(b)

FIG. 1. Phonon dispersions plotted along three high–symmetry lines. (a) Solid curves: Calculated phonon dispersion of UN. Open circles: Experimental phonon excitations of UN from Ref.[10]. (b) Theoretical phonon dispersion of UC.

that the use of the pole interpolation of self–energy

$$\Sigma(\omega) = \Sigma(\infty) + \sum_i \frac{V_i^\dagger V_i}{\omega - P_i} \tag{1}$$

allows us to replace the non–linear (over energy) Dyson equation by a linear Schroedinger–like equation in extended subset of "pole states":

$$\begin{pmatrix} \omega - H_0(\mathbf{k}) - \Sigma(\infty) - \epsilon_{\mathbf{k}j} & V^+ \\ V & \omega - P - \epsilon_{\mathbf{k}j} \end{pmatrix} \begin{pmatrix} \psi^{(e)}_{\mathbf{k}j} \\ \psi^{(a)}_{\mathbf{k}j} \end{pmatrix} = 0. \tag{2}$$

Here, only components $\psi^{(e)}_{\mathbf{k}j}$ describe one–electron excitations where the spectral content of each energy eigenvalue $\epsilon_{\mathbf{k}j}$ is determined by the matrix element $\langle\psi^{(e)}_{\mathbf{k}j}|\psi^{(e)}_{\mathbf{k}j}\rangle$ that is less than unity in general. The advantage of the present method is that the well developed machinery of standard electronic structure methods can be simply generalized to account for the dynamical self–energy effects. In particular, our successful applications on computation of phonon spectra in paramagnetic states of Mott–insulators such as NiO [2], UO_2 and PuO_2 [7], as well as in Pu [9] and Am [8] as representative systems with localized f–electrons have been made with the Hubbard 1 self–energy that exactly casts the form of the pole expansion in Eq. (1).

In the present work we extend this method to compute electron–phonon interactions for systems such as UC and UN whose f–electrons show itinerant behavior with $m^*/m_{LDA} \approx 4-12$. To capture this mass renormalization effect, we first make a fit to the self–energy obtained from the CTQMC, using a two–pole interpolation where the slope of the self–energy at zero frequency $d\Sigma(\omega)/d\omega|_{\omega=0} = 1 - m^*/m_{LDA}$ controls the electronic mass enhancement while the positions of the two poles P_i in Eq.(1) determine the transfer of the spectral weight from the quasiparticle band to the Hubard bands. Second, we assume that the f–electrons are rigidly bound to their ions so that there is no actual change in the self–energy, $\delta\Sigma(\omega)$, caused by ionic excursions from their equilibrium positions. Since the main contribution to electronic transport comes from the states near the Fermi surface, where quasiparticles are best described in terms of slave bosons, the neglecting of $\delta\Sigma(\omega)$ due to ion displacements corresponds to a rigid self–energy approximation. This is very similar to the famous rigid muffin–tin approximation (RMTA) [11], which has been successfully applied in the past to study electron–phonon interactions in transition metal materials [12, 13]. Therefore our use of rigid self–energy is expected to demonstrate a similar accuracy.

As a result, the electron–phonon scattering matrix element $g_{\mathbf{k}j\mathbf{k}+\mathbf{q}j'}$ can be evaluated using the electronic components $\psi^{(e)}_{\mathbf{k}j}$ that appear as solutions to Eq.(2), and the change of the ground state LDA potential, $\delta^q V_{LDA}$, computed for each phonon wave vector $\mathbf{q}$, i.e.

$$g_{\mathbf{k}j\mathbf{k}+\mathbf{q}j'} = \langle\psi^{(e)}_{\mathbf{k}j}|\delta^q V_{LDA}|\psi^{(e)}_{\mathbf{k}+\mathbf{q}j'}\rangle.$$

These matrix elements can be subsequently used for evaluating the EPI part of electrical and thermal resistivity similar to our previous applications on weakly correlated metals [4], where the corresponding Fermi surface integrals are now performed with "band structures" $\epsilon_{\mathbf{k}j}$ of Eq. (2) that acquire renormalizations due to correlations. By applying this theory, we have obtained a substantial reduction (by a factor of 3) in $\rho(T)_{EPI}$ for UC, while in UN the effect was marginal.

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