Electron correlation effects in $PuCoGa_5$ superconductor

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$PuCoGa_5$ is the first intermetallic superconductor containing plutonium, with the highest known critical temperature T_c= 18.5 K in actinide compounds [1]. Since plutonium belongs to strongly correlated electron systems, a possible mechanism involving spin-fluctuations was suggested [2]. The band structure calculations within LDA+U [3] and dynamical mean-field theory [4] found that electron correlations in 5*f* states on Pu play an important role.

A comparison between *ab initio* calculations [5] and inelastic X-ray measurements [6] showed that also phonon frequencies are strongly influenced by local Coulomb interaction *U*. The aim of this paper is to study the effect of *U* on electronic structure and phonon frequencies. The lattice constants, atomic positions, and interatomic forces were determined within the density functional generalized gradient approximation [7]. On-site Coulomb interactions, described by the Coulomb repulsion *U* and Hund's exchange *J* for the *f* states on Pu ions, were treated within the Hartree-Fock approximation. The calculations have been performed for nonmagnetic (NM), ferromagnetic (FM) and antiferromagnetic (AF) phases. In Fig. 1 we compare the electronic density of states of $PuCoGa_5$ for different phases with $UCoGa_5$ in the NM phase. AF and FM phases give better agreement with photoemision data [8]. Phonon frequencies were calculated using the direct method [9]. A schematic plot of phonon modes at the Brillouin center is presented in Fig. 2.

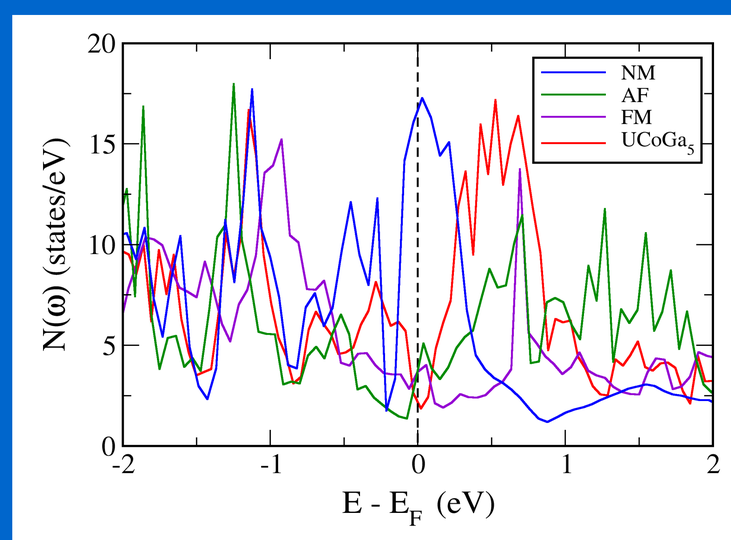

Fig. 1 Electron densities of states of $PuCoGa_5$ for NM (U = 0), AF and FM (U = 3 eV) states and $UCoGa_5$ for NM state (U =0).

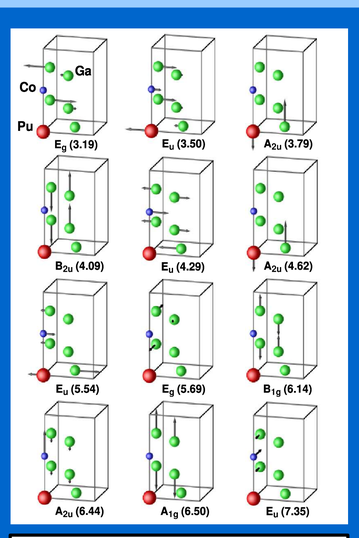

Fig. 2 Optical phonon modes at the Brillouin zone-center

The influence of Coulomb interaction *U* on phonon spectrum:

1. Stronger electron repulsion induces larger lattice constants and interatomic distances (see Tab. 1) -> overall phonon softening -> Fig. 3 (a),(b) and (c)
2. Electron correlations in *f* states on Pu changes charge distribution (partial localization) and electronic screening -> further softening of the lowest TO modes -> Fig. 4 -> hardening of LO phonon frequencies -> Fig. 5

	$PuCoGa_5$				$UCoGa_5$	
	NM	FM	AF	exp. [1]	NM	exp. [8]
a (Å)	4.197	4.230	4.236	4.232	4.240	4.234
c/a	1.595	1.641	1.630	1.603	1.582	1.588
z	0.304	0.312	0.311	0.312	0.306	0.305

Tab. 1 Lattice parameters

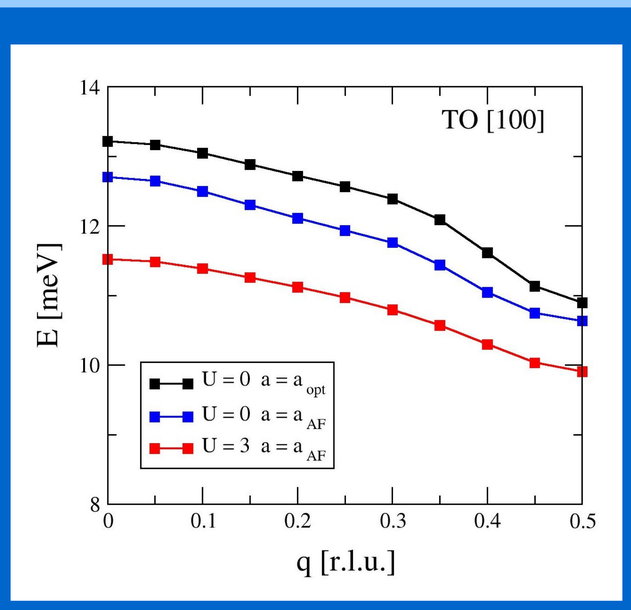

Fig. 4 TO mode in [100] direction

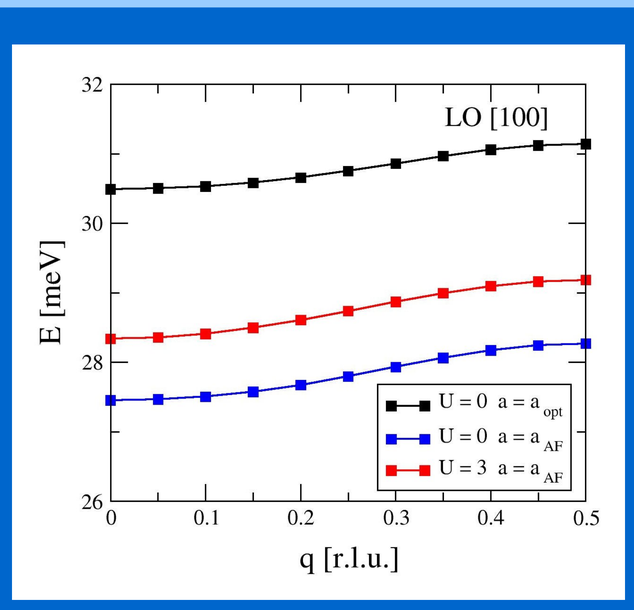

Fig. 5 LO mode in [100] direction

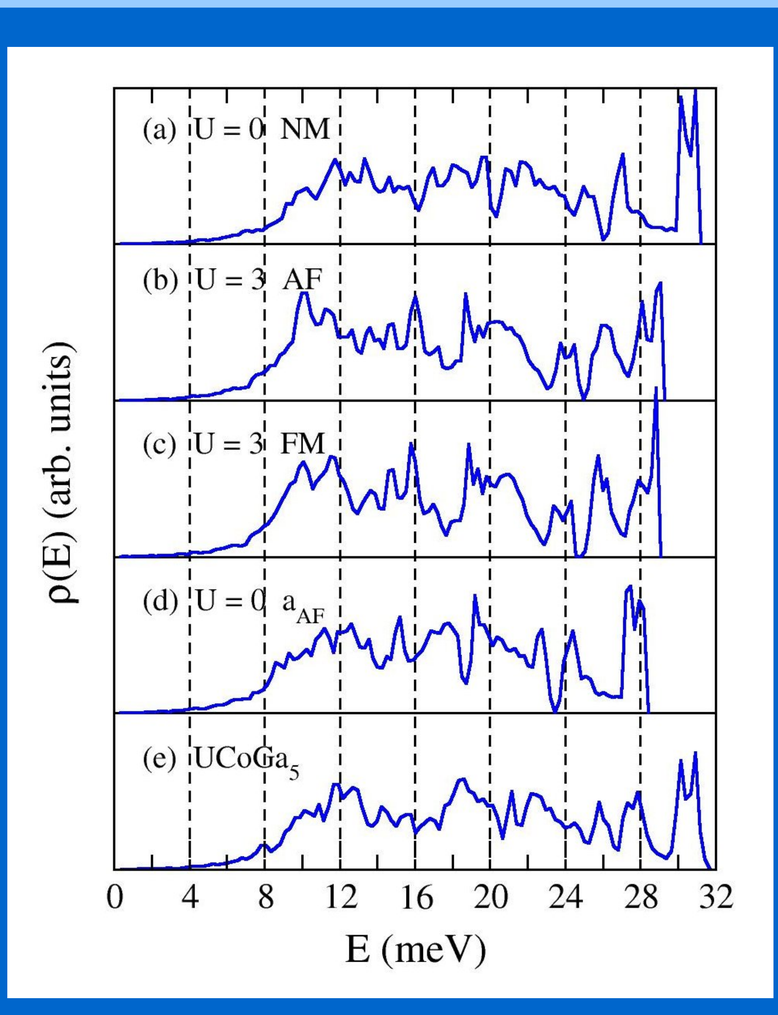

Fig. 3 Phonon densities of states in NM, AF, and FM state of $PuCoGa_5$ (a)-(d) and in NM state of $UCoGa_5$ (e).

$UCoGa_5$ has the same crystal structure as $PuCoGa_5$. Differences in phonon spectrum result from different electronic structures (Fig. 1). Phonon density of states [Fig. 3(e)] is broader than those of $PuCoGa_5$ in AF and FM states. Phonon dispersions of $UCoGa_5$ calculated in the NM state at $U = 0$ agree very well with the neutron measurement [10], indicating that the localized *f*-electron degrees of freedom are absent.

In conclusion, using *ab initio* calculations we demonstrated the effect of electronic structure on phonon spectra in $PuCoGa_5$ and $UCoGa_5$ crystals. The phonon spectra in the AF and FM phases of $PuCoGa_5$ are remarkably similar, indicating that electron localization plays an important role for phonons.

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