Influence of local Coulomb interactions on lattice dynamics 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]. The crystal structure of $PuCoGa_5$ is identical to that of Ce-based heavy fermion superconductors. Since plutonium belongs to strongly correlated f - electron systems, a possible mechanism involving spin-fluctuations was suggested [2]. Indeed recent LDA+U calculations demonstrated a strong effect on-site Coulomb interaction U at Pu ions on electronic band structure [3].

The aim of this paper is to study the effect of U on crystal structure and phonon frequencies. The lattice constants, atomic positions, and interatomic forces were determined within the density functional generalized gradient approximation (GGA) [4]. On-site Coulomb interactions, described by the intraorbital Coulomb repulsion U and Hund's exchange J element for the f states on Pu ions, were treated within the Hartree-Fock approximation. The crystal geometry optimization was done in the tetragonal 2x2x1 supercell with 28 atoms. The calculations have been performed for nonmagnetic (NM) and ferromagnetic (FM) phases. We found that the lattice parameters for $U > 0$ in FM phase agree better with the experimental data than those for NM phase with $U = 0$ (see Table 1). In Fig. 1 we compare the electronic density of states for $U = 0$ and 3 eV ($J = 0.7$ eV).

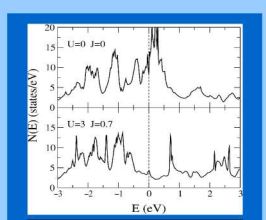

Fig. 1 The total electron densities of states for U=0 (upper) and U=3 eV (bottom)

Table 1.

crystal parameter	$U = 0$	$U = 3$ eV	experiment
a (Å)	4.207	4.304	4.271	4.203
c/a	2.595	2.420	2.456	2.503
z	0.304	0.311	0.308	0.305
V (Å^3)	117.8	124.7	124.7	121.0

The phonon frequencies were calculated using the direct method [5]. The Hellmann-Feynman forces were obtained for each nonequivalent atom at position n and i direction. The values of two diagonal elements of the force constants matrix xx and zz, computed for $U = 0$ and 3 eV, are shown in Table 2. All on-site force constants decrease with U as a result of the modification of crystal geometry (increase of average interatomic distance) and the charge redistribution due to Coulomb repulsion on Pu. The largest change is found for Ga atoms located in the Pu-Ga layers. There are also significant changes in both Pu force constants (about 15 %) and on Co, which decreases by about 25 %. Among all interatomic force constants, the most pronounced changes were found between Pu and Co atoms. A schematic plot of phonon modes at the Brillouin center is presented in Fig. 2. Phonon dispersion curves along high-symmetry directions are plotted in Figs. 3 and 4 for $U = 0$ and 3 eV, respectively.

Table 2

$n(xx)$-$n'(xx)$	Φ_{xx}		Φ_{zz}	
	$U=0$	$U=3$ eV	$U=0$	$U=3$ eV
Pu(1)-Pu(1)	-2.63	-3.79	-2.51	-3.64
Co(1)-Ga(1)	-0.34	-0.28	-7.46	-7.01
Ga(1)-Ga(1)	-2.35	-0.79	-2.49	-1.91
Ga(1)-Co(1)	-0.79	-3.59	-4.26	-7.10
Co(1)-Co(1)	-0.15	-0.11	-4.31	-4.25
Ga(1)-Co(2)	-1.11	-1.21	-3.31	-3.29
Pu(1)-Ga(2)	-4.06	-4.87	-3.20	-3.92
Co(1)-Co(2)	-0.22	-0.20	-4.12	-4.20
Pu(1)-Co(2)	-2.45	-1.87	-4.12	-4.08
Co(1)-Co(3)	-0.37	-0.43	0.21	0.18
Pu(1)-Ga(3)	-0.35	0.06	-0.95	-1.46

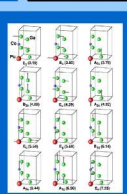

Fig. 2 Optical phonon modes at the Brillouin zone-center

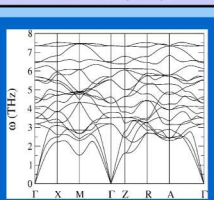

Fig. 3 Phonon dispersion curves for U = 0.

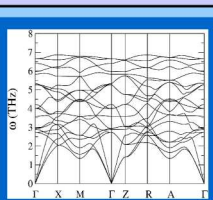

Fig. 4 Phonon dispersion curves for U = 3 eV.

The phonon density of states was computed by the summation of phonon modes over the entire Brillouin zone. In Fig. 5 we plot the total and partial phonon density of states (DOS) for each atom separately. The Coulomb interaction U leads to softening of phonon frequencies as expected from the analysis of the force constants. The largest modification (close to 30 %) is found for Ga atoms, vibrating at low frequencies. The Pu phonon DOS is slightly changed at frequencies around 5 THz. The highest optical modes, which correspond to Co movements, softens about 10 % due to changes in the on-site Co and Pu-Co force constants. The calculated phonon heat capacity for $U = 3$ eV is presented in Fig. 6.

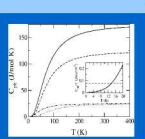

Fig. 6 Heat capacity

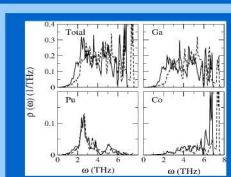

Fig. 5 Phonon total and partial density of states for U = 0 (dashed line) and U = 3 eV (full line).

In summary, we have studied the effect of local Coulomb interaction U on the crystal structure and lattice dynamics in $PuCoGa_5$. A better agreement between calculated lattice constants and the experimental data was found when Coulomb interactions were included. We have demonstrated that U changes significantly the force constant matrix elements, and influences lattice dynamics. This interplay between electronic distribution and lattice properties is likely to play an important role in the pairing mechanism.

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