Intermediate valence of Plutonium chalcogenides as evidenced by photoemission

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Intermediate valence is long standing field of the physics of condensed matter being part of strongly correlated electron systems. Usually this topic together with the physics of heavy fermions is observed in f electron systems such as rare earth and actinide compounds and alloys. Intermediate valence in actinides is still a rare case and the plutonium chalcogenides PuS, PuSe and PuTe seem to be the only well established materials of kind [1]. There, lattice constant, electrical transport data, magnetic susceptibility, specific heat and elastic properties have been used to establish these actinide compounds to be in the high pressure phase of the corresponding Sm chalcogenides which ate also intermediate valent. Thus e.g. PuTe at ambient pressure would electronically be in the same condition as SmTe under an applied pressure of 58 kbar [1].

Recently [2], UPS valence band photoemission spectra for PuSe were obtained with He I and He II excitation. The authors interpret their findings with localized f states and p band states originating in the Se 4p states and discard the intermediate valent condition of PuSe. However, the fit with the f final state multiplets ^{6}H, ^{6}F and ^{6}P (ground state Pu^{2+} ($^7F_0$)) yields only 3 lines below, but near E_F, whereas experimentally 4 lines are found. 4 eV below E_F another set of lines superimposes with the valence 4p band, but neglected by the authors [2], which can be associated with the f final state multiplets ^{5}I, ^{5}F, ^{5}G and ^{5}D, originating from Pu^{3+} (ground state ($^6H_{5/2}$)). Thus the Coulomb correlation energy in PuSe is about 4 eV in contrast with the one in the Sm chalcogenides of about 6 eV.

But at E_F there is one line more than can be associated with the f levels of Pu^{2+} alone. However, UPS with the He II line is still sensitive to 6d states, much less so with the He I line. We associate the strongest line in the UPS spectrum at E_F with the partially filled 6d band and we thus have a superposition or hybridization of 5f and 6d sates at E_F, a classical situation for intermediate valence. The optical reflectivity of PuSe yields a plasma resonance of the band-like 6d electrons just like in intermediate valent SmSe.

Thus the UPS results [2] also give clear evidence of intermediate valent PuSe.

References

[1] P. Wachter, F. Marabelli and B. Bucher, Phys. Rev. B 43 , 11136 (1991)

[2] T. Gouder, F. Wastin, J. Rebizant and L. Havela, Phys. Rev. Lett. 84, 3378 (2000)