Electronic Supplementary Material (ESI) for Chemical Science.

Origins of the Odd Optical Observables in Plutonium and Americium Tungstates

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Table S1. Crystallographic information for $PuW_2O_7(OH)(H_2O)$ at 100 K.

Compound	Pu
Formula Mass	755.76
Color and habit	Red, Plate
Space group	$P2_1/m$
a (Å)	5.8737(6)
b (Å)	8.6858(8)
c (Å)	7.0567(7)
a (°)	90
β (°)	105.408(2)
γ(°)	90
V (Å^3)	347.08(6)
Z	2
T (K)	100
λ (Å)	0.71073
Maximum 2θ (deg.)	30.57
ρB_{calcdB} (g cm^{-3})	7.212
μ(Mo Ka) (cm^{-1})	424.65
$R(F)$ for $F_o^2 > 2s(F_o^2)^a$	0.0236
$R_w(F_o^2)^b$	0.0569

Table S2. Bond lengths for $PuW_2O_7(OH)(H_2O)$ at 100 K.

PuW2O7(OH)(H2O)	
Pu(1)-O(2)	2.374(5)
Pu(1)-O(2)	2.374(5)
Pu(1)-O(3)	2.385(5)
Pu(1)-O(3)	2.385(5)
Pu(1)-O(4)	2.434(5)
Pu(1)-O(4)	2.434(5)
Pu(1)-O(5)	2.541(8)
Pu(1)-O(1)	2.755(7)
W(1)-O(4)	1.761(5)
W(1)-O(3)	1.764(5)
W(1)-O(2)	1.885(5)
W(1)-O(1)	1.961(4)
W(1)-O(2)	2.153(5)
W(1)-O(6)	2.197(5)

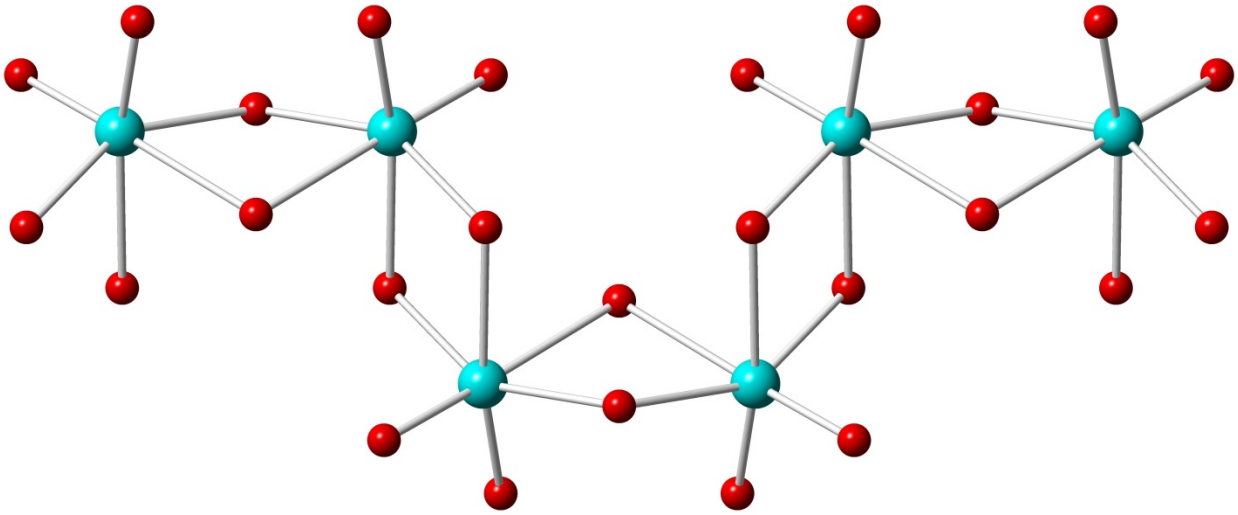

Figure S1. Ball-and-stick representation of infinite tungstate chains in MW2O7(OH)(H2O).

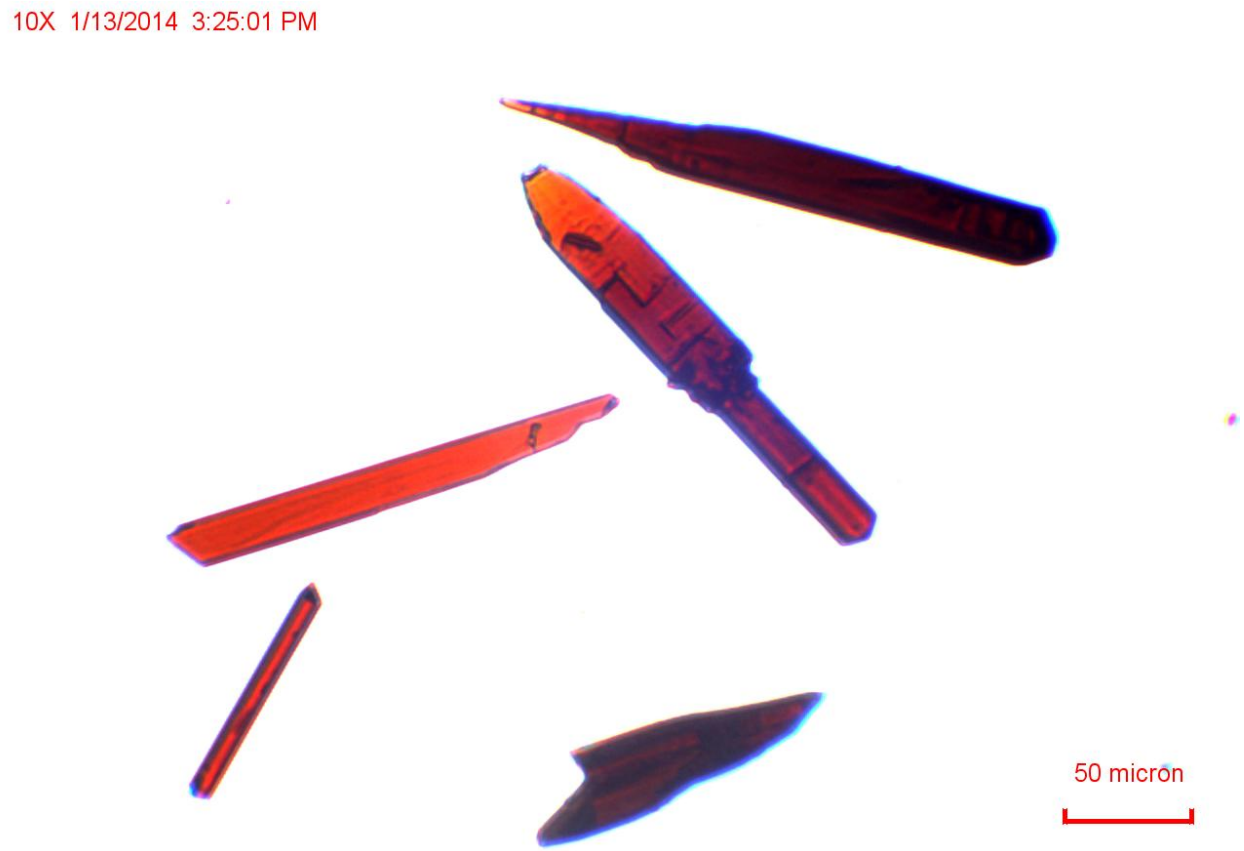

Figure S2. Photographs of crystals of PuW2O7(OH)(H2O).

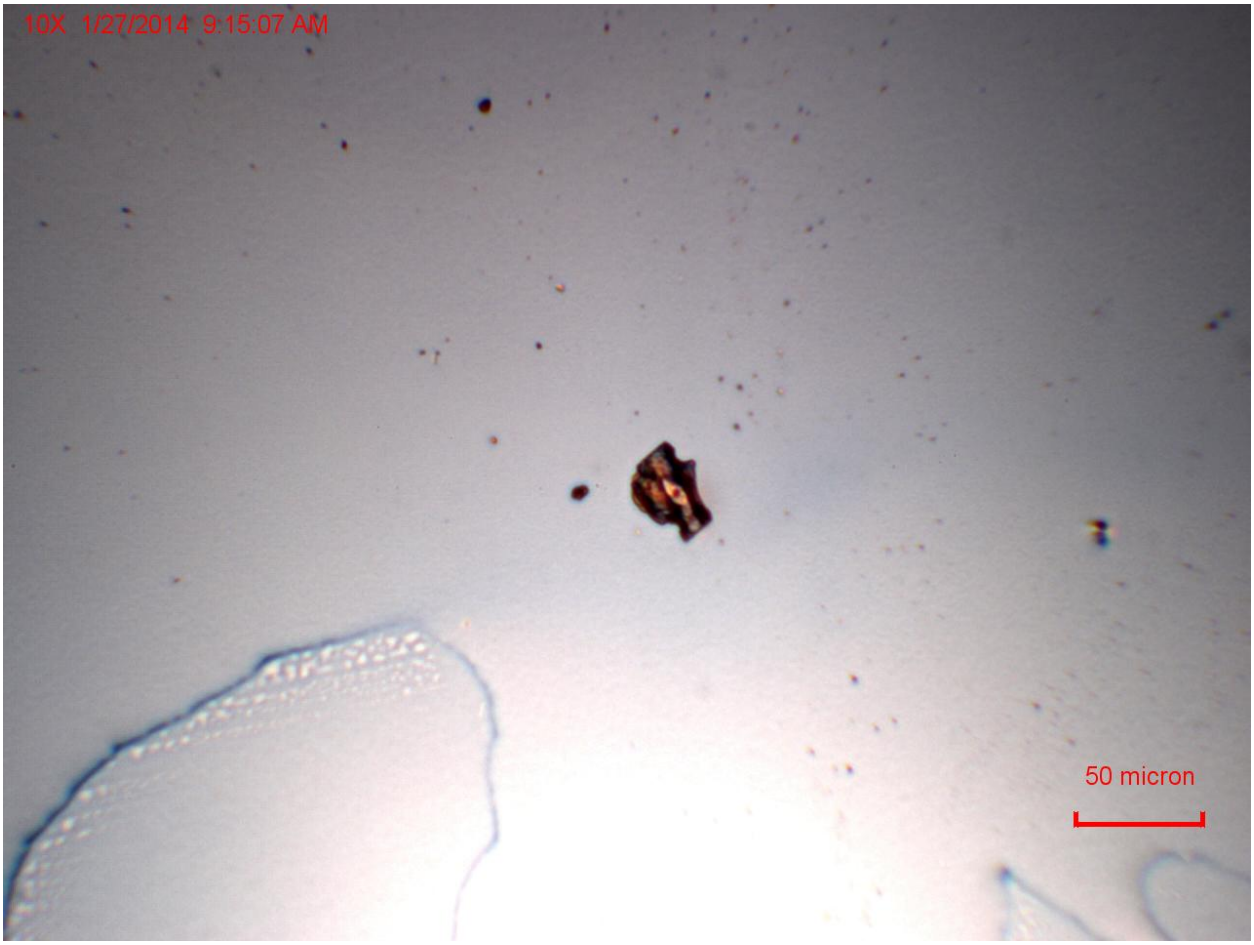

Figure S3. Photographs of crystals of AmWO4(OH).

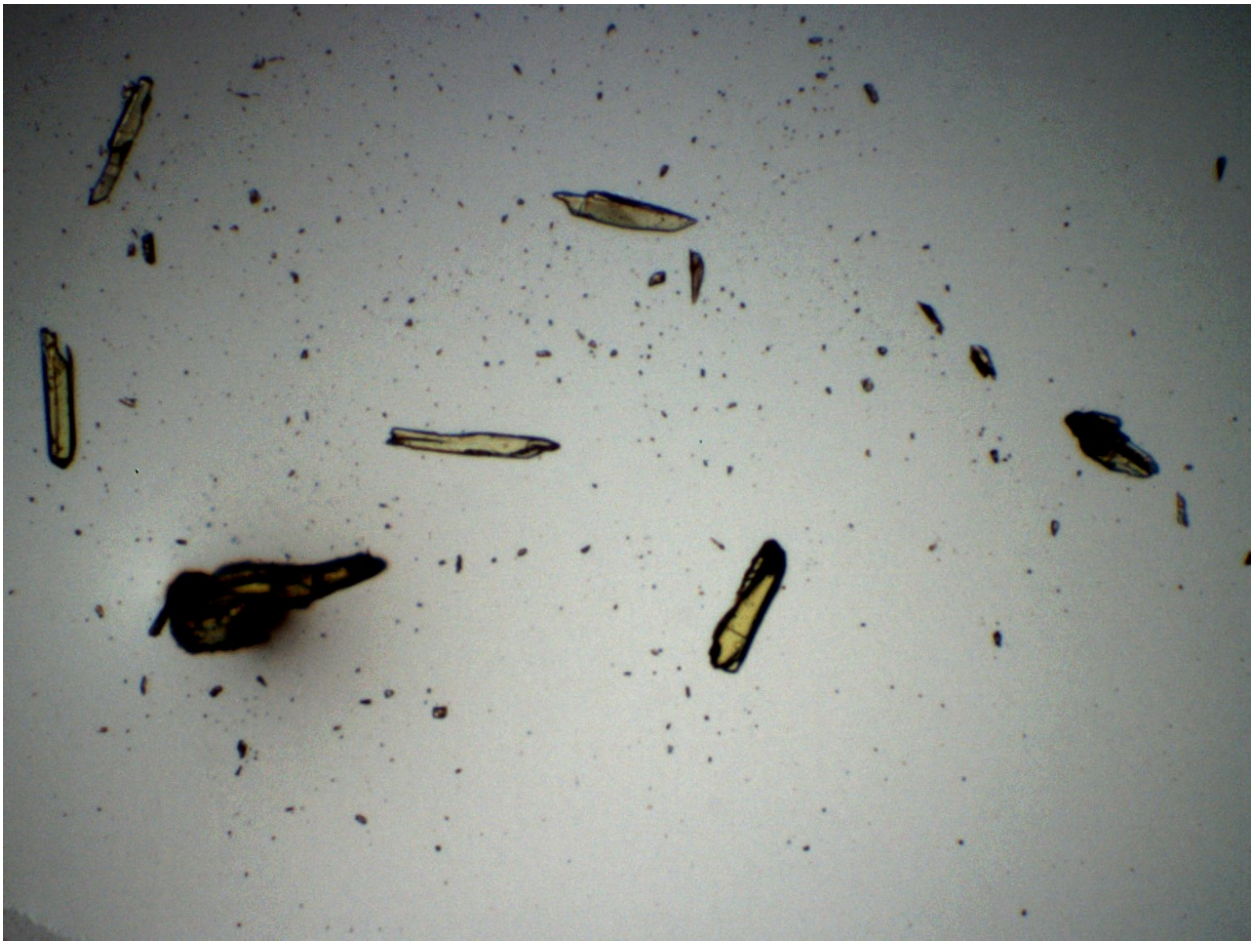

Figure S4. Photograph of crystals of $CeW_2O_7(OH)(H_2O)$.

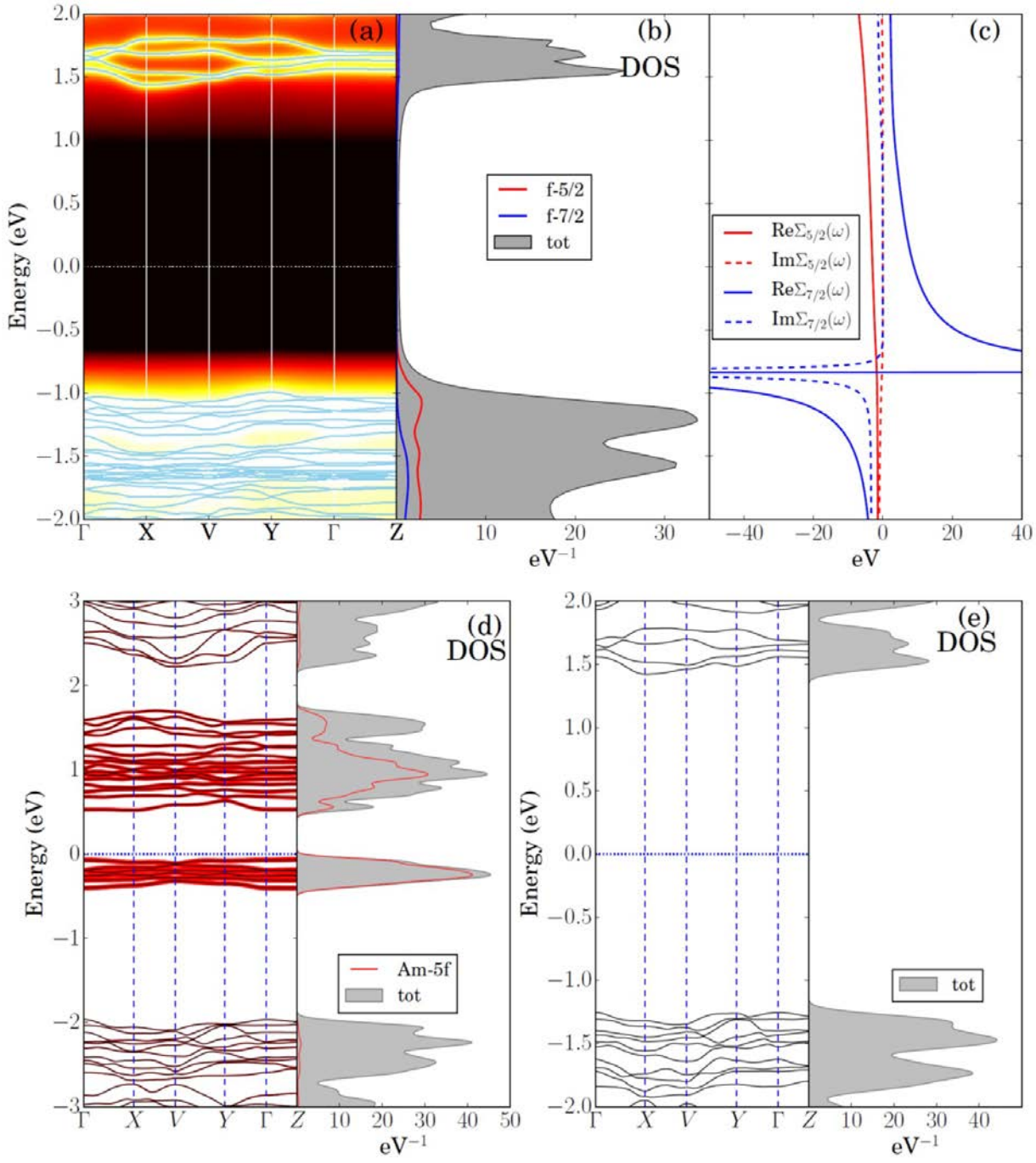

Figure. S5: Theoretical analysis spectral properties of $CsAm(CrO_4)_2$: (a) LDA+DMFT ARPES spectra computed at $T = 290\ K$ and corresponding quasi-particle bands. (b) Spectral contributions to the Am-5*f* 5/2 and 7/2 contributions to the DOS. (c) 5/2 and 7/2 components of the self-energy. (d) Bare unpolarized LDA bands and corresponding Am-5*f* 5/2 and 7/2 contributions to the DOS. (e) Modified LDA bands obtained by setting to 0 the hybridization between the 5*f* orbitals and their environment.

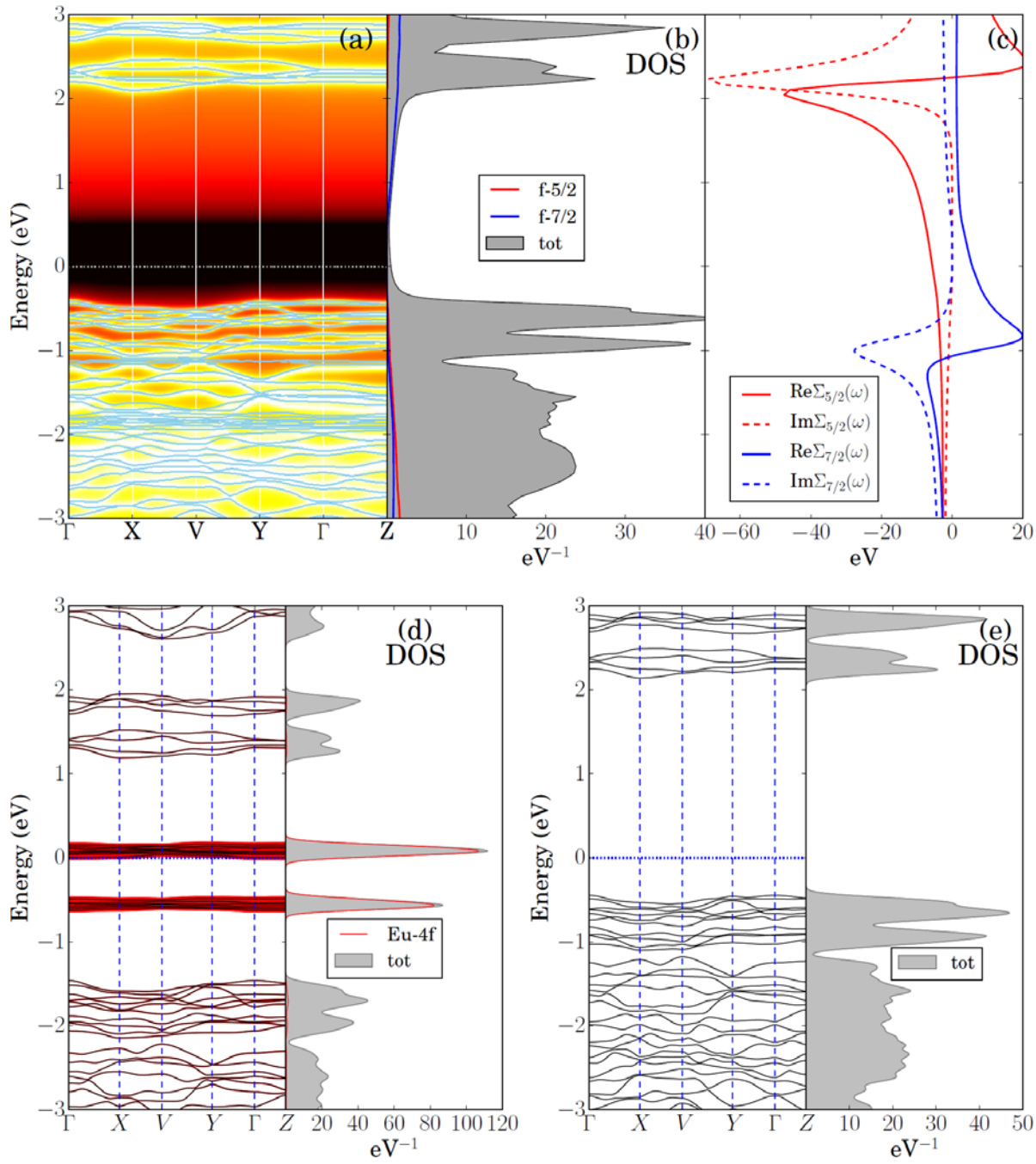

Figure. S6: Theoretical analysis spectral properties of $CsEu(CrO_4)_2$: (a) LDA+DMFT ARPES spectra computed at $T = 290\ K$ and corresponding quasi-particle bands. (b) Spectral contributions to the Eu-4*f* 5/2 and 7/2 contributions to the DOS. (c) 5/2 and 7/2 components of the self-energy. (d) Bare unpolarized LDA bands and corresponding Eu-4*f* 5/2 and 7/2 contributions to the DOS. (e) Modified LDA bands obtained by setting to 0 the hybridization between the 4*f* orbitals and their environment.

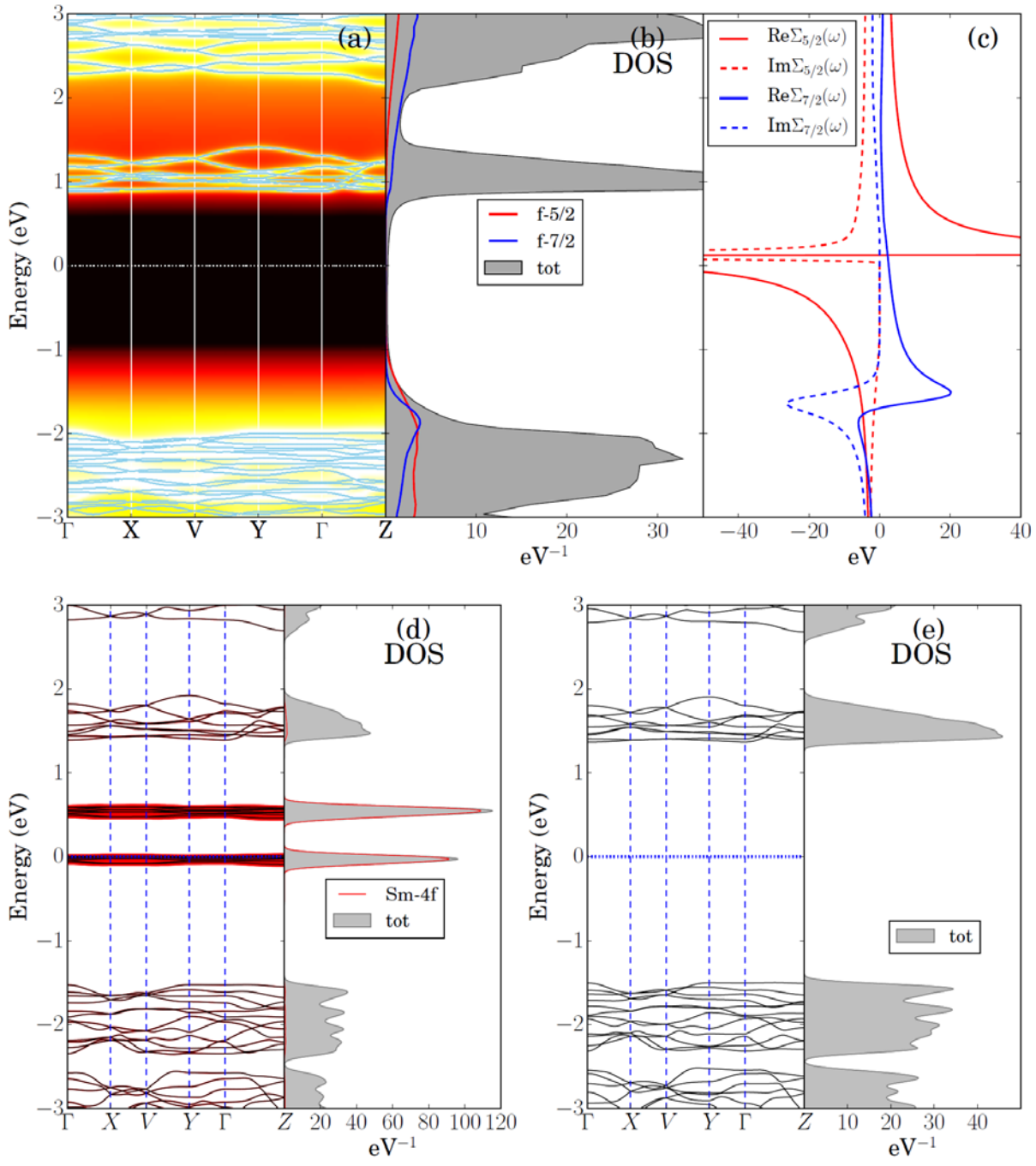

Figure. S7: Theoretical analysis spectral properties of $CsSm(CrO_4)_2$: (a) LDA+DMFT ARPES spectra computed at $T = 290\ K$ and corresponding quasi-particle bands. (b) Spectral contributions to the Sm-4*f* 5/2 and 7/2 contributions to the DOS. (c) 5/2 and 7/2 components of the self-energy. (d) Bare unpolarized LDA bands and corresponding Sm-4*f* 5/2 and 7/2 contributions to the DOS. (e) Modified LDA bands obtained by setting to 0 the hybridization between the 4*f* orbitals and their environment.