Supplementary Information:

First-Principles Study of Metal and Ligand Substitution Effects on EUV Absorption and Electron Energy Loss

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A.1. Structural models and relaxation: Geometry optimization were performed with the projector augmented wave method, as implemented in VASP. Exchange and correlation effects were treated in the generalized gradient approximation (GGA) using the Perdew Burke Ernzerhof functional (PBE).[1,2] Our relaxation protocol consists in a full structure relaxation with the conjugated gradient method until Hellman-Feynman forces on all atoms were smaller than 0.01 eV/Å with a calculation precision energy equal to 10^{-4} eV.[3] K-point integration of the Brillouin zone was set automatically with a KSPACING parameter of 0.5. A plane wave basis was used for wave function expansion, with a kinetic energy cutoff of 500 eV (greater than 1.3 × default cutoff as recommended) to ensure the convergence of total energy during the self-consistent calculation loop.[4]

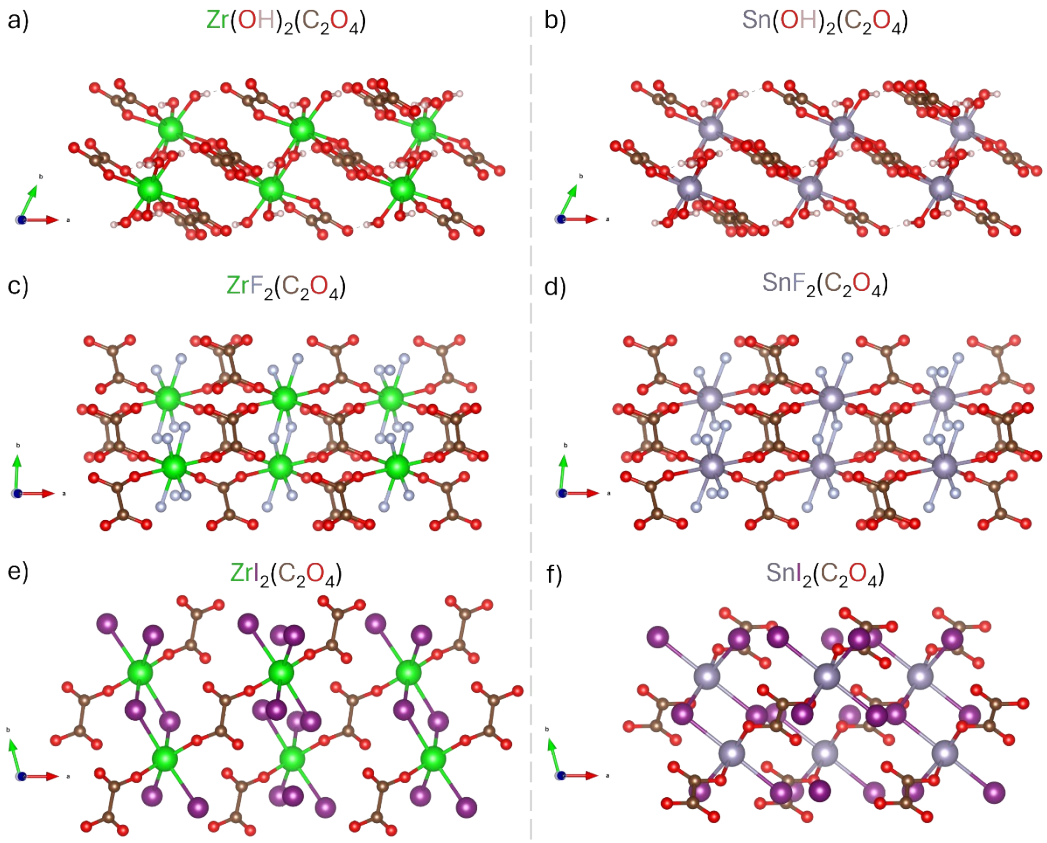

Figure S1. $Zr(OH)_2(C_2O_4)$ (from the experimental structure), $ZrF_2(C_2O_4)$, $ZrI_2(C_2O_4)$, $Sn(OH)_2(C_2O_4)$, $SnF_2(C_2O_4)$ and $SnI_2(C_2O_4)$, fully relaxed structure using VASP.[45]

Table S1. Lattice parameters and metal-ligands bond lengths in the $ML_2(C_2O_4)$ systems after geometry optimization with the VASP package (as described).

	a (Å)	b (Å)	c (Å)	α (°)	β (°)	γ (°)	Metal-ligands (Å)
$Zr(OH)_2(C_2O_4)$	12.68	6.65	5.69	90.00	90.00	64.19	2.12 / 2.11
$ZrF_2(C_2O_4)$	11.22	6.85	5.98	90.00	90.00	87.54	2.20 / 2.08
$ZrI_2(C_2O_4)$	13.16	8.70	7.66	90.00	90.00	75.35	3.01 / 2.93
$Sn(OH)_2(C_2O_4)$	12.65	6.68	6.06	90.00	90.00	63.33	2.17 / 2.09
$SnF_2(C_2O_4)$	11.34	6.61	5.97	90.00	90.00	84.19	2.19 / 2.04
$SnI_2(C_2O_4)$	9.95	7.42	14.28	90.00	90.00	74.71	2.94 / 2.92
$Sn[I->F]_2(C_2O_4)$	9.95	7.42	14.28	90.00	90.00	74.71	2.94 / 2.92

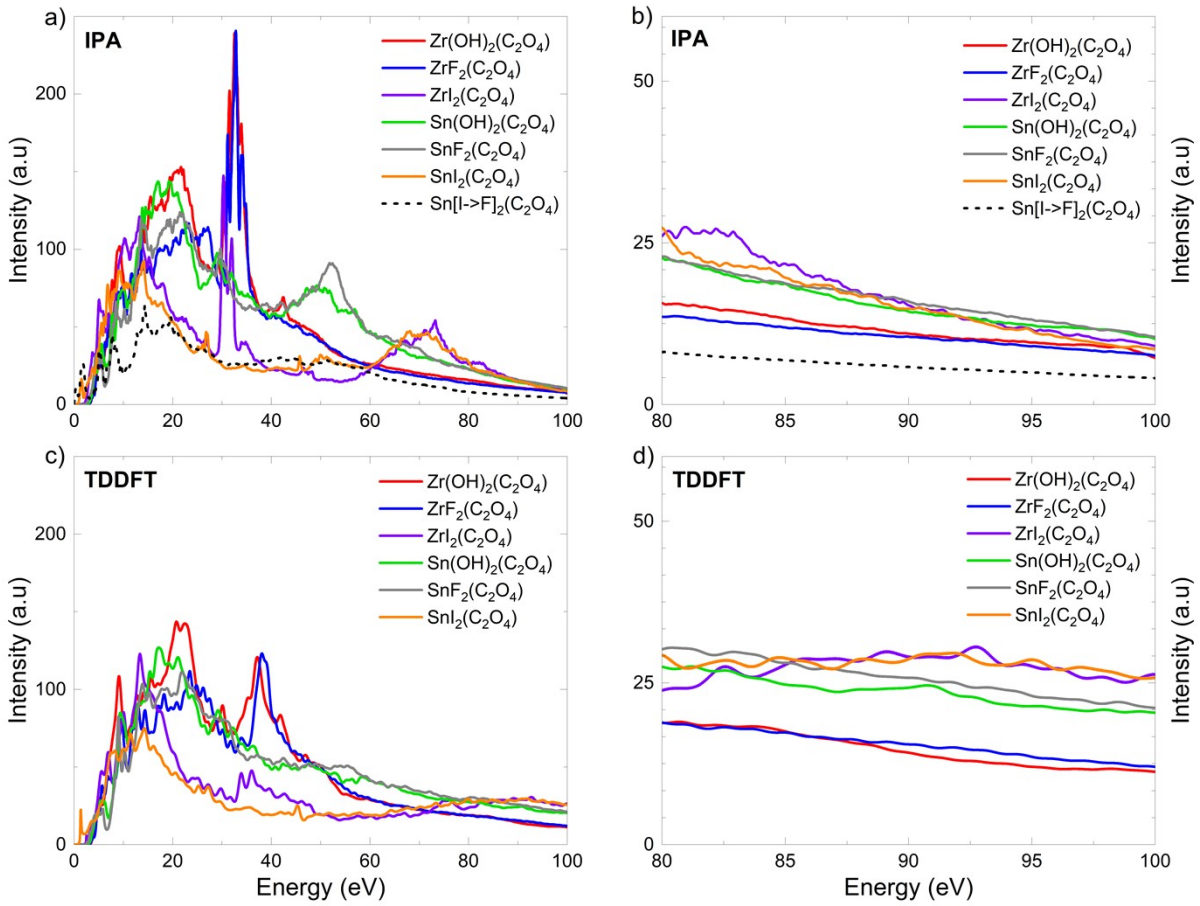

Figure S2. Calculated optical absorption spectra of the $Zr(OH)_2(C_2O_4)$, $ZrF_2(C_2O_4)$, $ZrI_2(C_2O_4)$, $Sn(OH)_2(C_2O_4)$, $SnF_2(C_2O_4)$ and $SnI_2(C_2O_4)$ model systems, in IPA a) from 0 to 100 eV and b) from 70 to 100 eV, and TDDFT c) from 0 to 100 eV and d) from 70 to 100 eV. a) and b) Dashed line $Sn[I->F]_2(C_2O_4)$: substitution of I atoms by F atoms in the $SnI_2(C_2O_4)$ structure (longer Sn-F bond than in $SnF_2(C_2O_4)$), highlighting the decreases of optical absorption when the orbital overlap is reduced

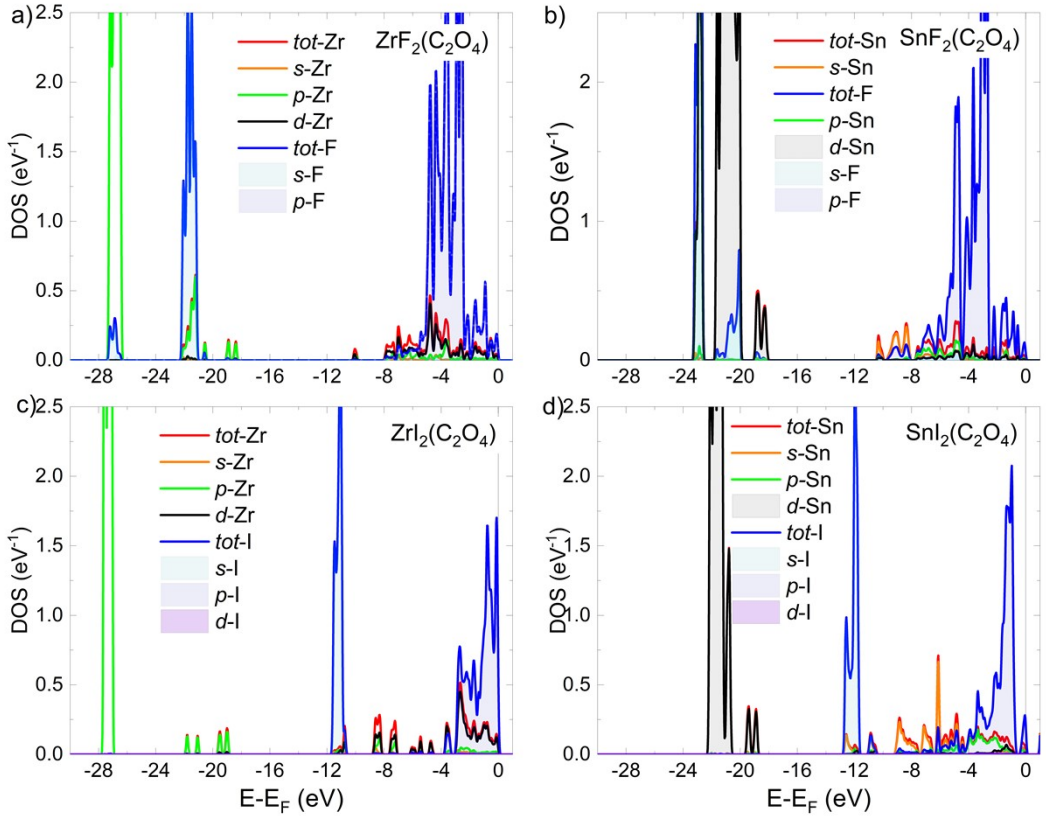

Figure S3. Partial density of states (pDOS) of a) $ZrF_2(C_2O_4)$, b) $SnI_2(C_2O_4)$, c) $ZrI_2(C_2O_4)$ and d) $SnI_2(C_2O_4)$, calculated with the WIEN2k code.

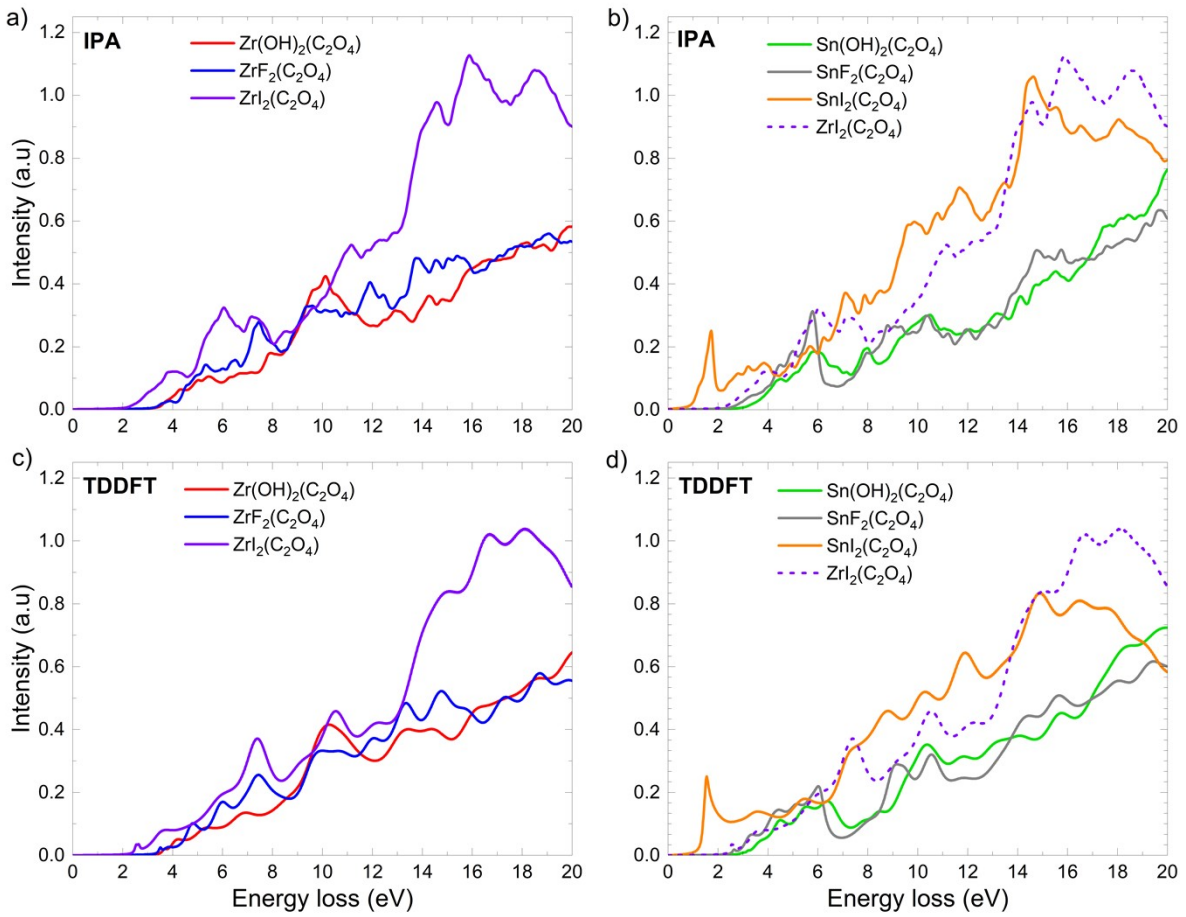

Figure S4. Calculated energy loss function (ELF) from 0 to 20 eV of a) $Zr(OH)_2(C_2O_4)$, $ZrF_2(C_2O_4)$ and $ZrI_2(C_2O_4)$ in IPA b) $Sn(OH)_2(C_2O_4)$, $SnF_2(C_2O_4)$ and $SnI_2(C_2O_4)$ in IPA c) $Zr(OH)_2(C_2O_4)$, $ZrF_2(C_2O_4)$ and $ZrI_2(C_2O_4)$ in TDDFT d) $Sn(OH)_2(C_2O_4)$, $SnF_2(C_2O_4)$ and $SnI_2(C_2O_4)$ in TDDFT. For an easier comparison the ELF of $ZrI_2(C_2O_4)$ is added (for both theories) to the Sn containing systems in dashed line.

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