## **Supporting Information**



**Figure S1.** A representation of the translational quadruple phenyl embrace TQPE in **1**. Symmetry code: (a) = 1 - x, 1/2 + y, 1/2 - z.



Figure S2. View along the crystallographic b axis of the crystal packing of 1 showing the alternating arrangement of the cations (C) and anions (A).



**Figure S3**. Perspective drawing along the crystallographic *a* axis of a fragment of the double zigzag chain of **3**. The thermal ellipsoids are drawn at the 50% probability level and the hydrogen atoms are omitted for clarity. Symmetry code: (a) = x - 1, y, z; (b) = -x + 1, -y, -z + 1; (c) = -x, -y, -z + 1; (d) = x + 1, y, z.



**Figure S4.** Perspective view in the plane *bc* of two neighboring chains of **3** showing the orientation of the vectors (*v1* and *v2*) of the ribbon-like structure. Hydrogen bonds are drawn as broken lines. Symmetry code: (j) =  $x - \frac{1}{2}$ ,  $-y + \frac{1}{2}$ ,  $z - \frac{1}{2}$ ; (l) = -x, -y, -z + 1.



Figure S5. Schematic representation of the 3D structure formed *via* hydrogen bonds (broken lines) between the bimetallic chains of 3. The phen ligands and the hydrogen atoms were omitted for the sake of clarity. Symmetry code: (g) = x - 1/2, y + 1/2, z - 1/2.



Figure S6. Heat capacity measurements on a pellet of 3 at very low temperatures.