Thermoelectric properties of n-type PbS nanocrystals doped with two-dimensional lead bromide perovskites

Marios E. Trantafyllou-Rundell,[a,b] Christine Fiedler[b], Constantinos C. Stoumpos,[a] Maria Ibáñez[b]

[a]Department of Materials Science and Technology, University of Crete, Heraklion 70013, Greece

[b]Institute of Science and Technology Austria, Klosterneuburg 3400, Austria

Thermoelectric materials have been highly-sought-after materials since they promise to convert wasted heat energy into useful electrical power, using the so called Seebeck effect.[1,2].The efficiency of a thermoelectric material is expressed by the figure of merit zT which is defined as $zT = S^2\sigma T/(\kappa_e+\kappa_L)$ where S is the Seebeck coefficient, σ is the electric conductivity, and κ_e and κ_L are the electron and lattice thermal conductivity, respectively. Lead chalcogenides have attracted most of the attention in the past decades, as they fulfill the requirements for achieveing high figures-of-merit (high σ, small κ) while the temperature of optimal operation lies in the few hundred degrees Celsius, an ideal temperature to convert heat from combustion engine exhausts. PbS-based materials have attracted much attention for thermoelectric power generation due to their low-cost and earth-abundant features.

In this work we crystallized 2D metal halide perovskites on top of n-type PbS nanoparticles in order to dope (n-type) the PbS into PbS/perovskite composites. $(HA)_2PbBr_4$ perovskite crystals (where HA stands for the protonated monoanionic form of hexylamine) dissolved in a suitable polar solvent were intermixed with PbS nanoparticles dispersed in the same solvent under controlled N_2 atmosphere. The addition of nonpolar solvent leads to the rapid recrystallization of the perovskite on the surface of the PbS nanocrystals. Thermal annealing and coalescence using Spark Plasma Sintering (SPS) conditions, followed by appropriate processing led to dense pellets of the composite material. Overall, halide perovskites succeed in increasing carrier concentration and electrical conductivity of PbS. Thereby, the power factor is improved (~15 μW cm^{-1} K^{-2}) pushing the zT value up to ~ 0.8 at 850K. (Fig 1.)

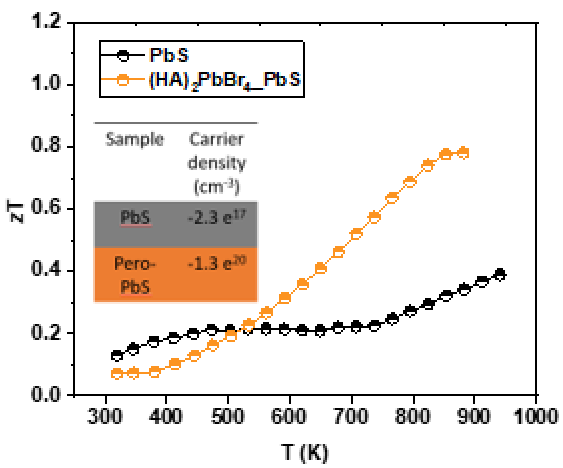

Figure 1. zT of the PbS pellet before and after the perovskite.

[1] Zhao, L.-D. *et al.* High Performance Thermoelectrics from Earth-Abundant Materials: Enhanced Figure of Merit in PbS by Second Phase Nanostructures. *J. Am. Chem. Soc.* **133**, 20476–20487 (2011).

[2] Hu, S., Ren, Z., Djurišić, A. B. & Rogach, A. L. Metal Halide Perovskites as Emerging Thermoelectric Materials. *ACS Energy Lett.* **6**, 3882–3905 (2021).