KN1200

Reactive deposition of nitrides and oxide coatings for thermoelectrics

Per Eklund

Linkoping University, Linkoping, Sweden

perek@ifm.liu.se

Thermoelectric devices have the potential to contribute to energy harvesting in society by directly converting heat into electricity or vice versa. However, the conversion efficiency of thermoelectric devices of today is limited. The critical material-dependent parameter is the figure of merit ($ZT = S^2T/\rho\kappa$, where ρ is the electrical resistivity, S is the Seebeck coefficient and κ is the total thermal conductivity). In this keynote lecture, I present an overview of our experimental and theoretical investigations of CrN-, ScN-, and $Ca_3Co_4O_9$-based systems by reactive magnetron sputtering. We have introduced a two-step sputtering/annealing method for the formation of highly textured virtually phase-pure $Ca_3Co_4O_9$ thin films by reactive co-sputtering from Ca and Co targets followed by an annealing process at 730 °C under O_2-gas flow. The thermally induced phase transformation mechanism was investigated by in-situ time-resolved annealing experiments using synchrotron-based 2D x-ray diffraction as well as ex-situ annealing experiments and standard lab-based x-ray diffraction. By tuning the proportion of initial CaO and CoO phases during film deposition, the method enables synthesis of $Ca_3Co_4O_9$ thin films as well as Ca_xCoO_2. ScN thin films exhibit an anomalously high power factor (S^2/ρ) for transition metal nitrides of $2.5\text{-}3.3\times10^{-3}$ W/mK^2 at 800 K [1]. We have explained this result, from first-principles calculations [2], by nitrogen vacancies generating an asymmetric sharp feature in the density of states which allows low electrical resistivity with relatively large S. However, ScN has high thermal conductivity, thus its ZT is low (~0.2). To reduce lattice thermal conductivity, potential strategies are nanostructuring, alloying or nanoinclusion formation. Superlattice structures and alloying with heavy elements have been demonstrated to reduce thermal conductivity [3], and our experimental results demonstrate that alloying with Cr approaches the high ZT value of CrN [4]. Our results demonstrate that pure CrN exhibits a high power factor, 1.7×10^{-3} Wm^{-1}K^{-2} at 720 K, enabled by a high electron concentration thermally activated from N vacancies and corresponding to an estimated thermoelectric figure of merit ZT as high as ~0.5.

Keywords
reactive sputtering
thermoelectrics