First-principles approach to calculate entropy in magnetocaloric materials.

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The increasing interest in the application of magnetocaloric materials for magnetic cooling devices has led to an intensive search for new materials with a more attractive performance to cost ratio. Yet, many magnetocaloric materials display complex behaviours, which are not fully comprehended. High-throughput studies based on first-principles calculations can play a crucial role to detect new magnetocaloric material while increasing the insight on these materials. However, to identify systems of interest in a large body of data, screening parameters are required and must be carefully chosen considering a balance between accuracy and cost of the calculations. A key quantity to characterize the performance of these systems is the entropy variation between two magnetic phases. In this work, electronic, structural and magnetic properties of bulk FeRh and Gd have been studied from first principles aiming to find a reliable non-tailored approach to determine the entropy variation of the magnetocaloric effect being obtained good agreement with experimental results for the total entropy.

In case of FeRh the electronic (S_{ele}), lattice (S_{lat}), and magnetic (S_{mag}) entropy contributions have approximately the same order of magnitude and the same sign. For Gd, we find that besides S_{mag}, it is important to include the contribution of S_{ele} for an accurate estimation of the total entropy variation.

Furthermore, the calculated entropy contributions for FeRh suggest that the magnetic subsystem drives the metamagnetic transition. Taking into consideration the used model, the adiabatic magnon model, there is a strong indication that small magnetic fluctuations play an important role in the magnetic transition. Also, it is found for FeRh that the Debye model cannot accurately predict S_{lat} due to the existence of soft vibrational modes on the phonon spectra.

References:

[1] Martinho Vieira, R., Eriksson, O., Bergman, A., Herper, H. C. (2020) Journal of Alloys and Compounds, 157811.
[2] Cooke, D. W., Hellman, F., Baldasseroni, C., Bordel, C., Moyerman, S., Fullerton, E. E. (2012) Physical Review Letters, 109(25), 255901.
[3] Gschneidner Jr, K. A., Pecharsky, V. K. (2006). Journal of Rare Earths, 24(6), 641-647.

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