Surfing Between Structure and Spin State: High-Pressure Studies of Spin Crossover Compound

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Abstract

Spin crossover (SCO) materials are attracting growing interest for many applications due to their ability to switch reversibly between high spin (HS) and low spin (LS) states under numerous external stimuli such as temperature, light or pressure. According to this last stimulus and due to their ability to switch reversibly between HS and LS associated with a high entropy variation, SCO complexes have been recently identified as potential candidates for solid-state barocaloric refrigerant[1,2] (for the cooling systems of the future). Within this context, designing molecular-based spin transition materials that are optimal for barocaloric application requires a thorough understanding of SCO behavior under extreme conditions.

In this work, we investigate a spin-crossover material as a model system to better understand the effects of pressure on functional materials. Our study focuses on its thermodynamic behavior though the construction of a pressure-temperature phase diagram. To achieve this, we employ advanced X-Ray diffraction or absorption techniques, which enable us to probe structural changes associated with spin transition under pressure and temperature. By systematically exploring a wide range of pressures and temperatures with small steps, we identify phase boundaries and characterize the coexistence of distinct spin states. The resulting phase diagram contributes to a deeper understanding of how external pressure influences phase stability and transition mechanisms in such systems, offering a broader perspective on their physical properties and potential technological relevance.

Beyond the targeted SCO properties (such as $T_{1/2}$ and transition abruptness), identifying suitable candidates for barocaloric application requires a thorough understanding of the properties and stability of the different phases under pressure. The results presented here therefore illustrate one of the essential steps that must be undertaken before considering a spin crossover material for barocaloric applications.

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(2) Lloveras, P.; Tamarit, J.-L. Advances and Obstacles in Pressure-Driven Solid-State Cooling: A Review of Barocaloric Materials. *MRS Energy Sustain.* **2021**. https://doi.org/10.1557/s43581-020-00002-4.

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