'Invar' behavior in a chiral helimagnet

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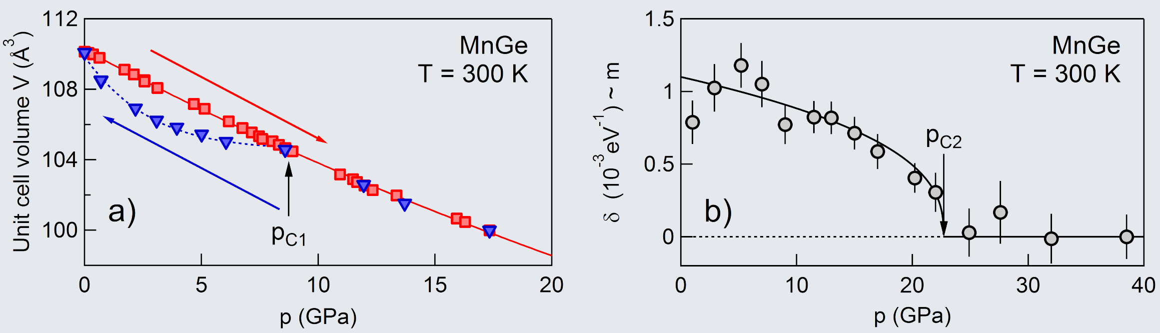
The history of the 'invar' behavior traces back to the end of XIXth century, when C.E. Guillaume synthetized and studied Fe-Ni ferromagnetic alloys, characterized by an invariant lattice parameter around room temperature (hence the term 'invar'). Since its discovery, several models have been developped to understand this peculiar phenomenon and -let apart technical details- magneto-elastic coupling is unanimously acknowledged as an essential ingredient, explaining why different properties of the materials, such as the atomic volume V, local magnetic moment m or bulk modulus B can be affected. One of the most popular theory of the 'invar' effect is due to P. Weiss who introduced the so-called 2γ-model, where a high-spin (HS) state with large atomic volume and magnetic moment competes with a low-spin (LS) metastable state with reduced volume and moment. Here, a thermal activation of the LS state counteracts lattice thermal expansion (LTE). However, as pointed out by D.I. Khomskii and F.V. Kusmartsev (KK), this simple mechanism cannot explain the cancelation of LTE over large temperature intervals as experimentally observed. It is therefore necessary to feed additional ingredients into the model -one of which being intersite elastic coupling between minority spin states- and verify their validity on real systems. In this context, chiral magnet MnGe appears as a good playground since neutron powder diffraction (NPD) has revealed a HS-LS transition at low temperature, within its helimagnetic state.



Here, we have made use of X-ray powder diffraction (XPD) and emission spectroscopy (XES) to study the pressure-dependence of the unit cell volume V (**Fig. a**) and Mn local moment m (**Fig. b**) of MnGe. Our initial aim was to ascertain the existence of an intermediate LS state at T = 300 K, *i.e.* in the paramagnetic regime where long-range magnetic order is absent. As seen on **Fig. a**, V describes a remarkable hysteresis loop upon pressure cycling. This loop opens upon decompression at $p_{C1} \approx 7$ GPa, which corresponds fairly well to the HS-LS critical pressure of ≈ 6 GPa determined by NPD. Interestingly, the existence of such elastic irreversibilities perfectly supports the idea of KK that the nucleation of low volume LS regions induces local strains which couple over finite distances through the sample. In other words, they build up a macroscopic energy barrier that has to be overcome to transform the system back to its initial HS state. We have developped a thermodynamic model of the spin-lattice transition which accounts for the observed V = f(p) curve. In parallel, a challenging XES experiment has allowed us to follow the pressure-dependence of the local Mn moment up to ≈ 40 GPa. As shown in **Fig. b**, m collapses at $p_{C2} \approx 23$ GPa and leads to a non-magnetic state. This is another way of verifying the existence of an intermediate LS state and assessing its metastable nature.

In summary, our study establishes MnGe as the first known example of 'invar' chiral magnet. Moreover, our findings *directly* verify the theoretical prediction of KK that intersite coupling must play an important role in the appearance of 'invar' anomalies, thus updating common viewpoints on the physics at play in these systems.

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