Strain Tuning of Correlated and Frustrated Magnetic Materials

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With our department's pioneering development of piezoactuator-driven uniaxial pressure cells, we can now manipulate quantum materials in ways that cannot be achieved by chemical methods. In this report, we describe how we use the new experimental tools to explore the stress and strain control of correlated magnets, which realize fascinating phenomena arising from electronic correlations and topology. By exploiting the precise control of uniaxial pressure, we can break or restore the crystal lattice symmetry to selectively tune the underlying competing magnetic interactions. With our thermodynamic experiments under these extreme conditions, we aim to address major unresolved questions in the field, such as the properties of a maximally frustrated magnet, and to discover new quantum states and their properties.

Correlated quantum magnets are a central focus in condensed matter research because of the fundamental quantum physics they reveal and their putative practical applications. In this context, both materials that exhibit magnetic order and those that lack magnetic order despite strong interactions are equally intriguing and means to continuously tune between one and the other are highly desired. Our current understanding suggests that this can be achieved by controlling the strength of the magnetic frustration. However, the precise control of the frustration strength in experiments is a major challenge in the field. Achieving this high-precision control of frustration and competing interactions has the potential to significantly advance the field by bridging the gap between theoretical model calculations and experimental phase diagrams.

Uniaxial pressure provides an excellent opportunity to achieve this high-precision tuning. Consequently, we focused our efforts during this census period on using uniaxial pressure to tune interactions and frustration in correlated magnets. To achieve this, we adapted methods established in our department's successful research on the unconventional superconductor Sr_2RuO_4 under large strains (see **PQM_04_Noad** and **PQM_02_Nicklas**). In particular, high-precision thermodynamic probes, such as the AC elastocaloric effect [1] and the stress-strain relationship [2], are essential for the study of correlated magnetism under strain. We have successfully applied these techniques to a broad range of materials, including soft, fragile, and insulating materials. While our primary focus is on studying the effects of frustration in magnetic insulators, we also study other correlated systems to improve our conceptual understanding of the elastocaloric effect and the stress-strain relationship as well as of the interplay of correlated electronic and lattice degrees of freedom. As we will illustrate with specific examples, our work to date has laid an important foundation for future research on correlated, frustrated magnets under large strains.

Strain control of the 'super'-heavy fermion YbPtBi

In an important piece of work, we extended the study of the elastocaloric effect – an exciting new thermodynamic probe for quantum materials – to a material with very strong correlations. For this, we chose to study the 'super'-heavy fermion YbPtBi, a compound that is famous for its proposed record-high specific heat at low temperatures, the origin of which has remained a great mystery since its discovery. One of the possible explanations brought forward attributed the large electronic mass to the high degeneracy of crystal-electric field levels in this cubic heavy fermion below the Kondo temperature.

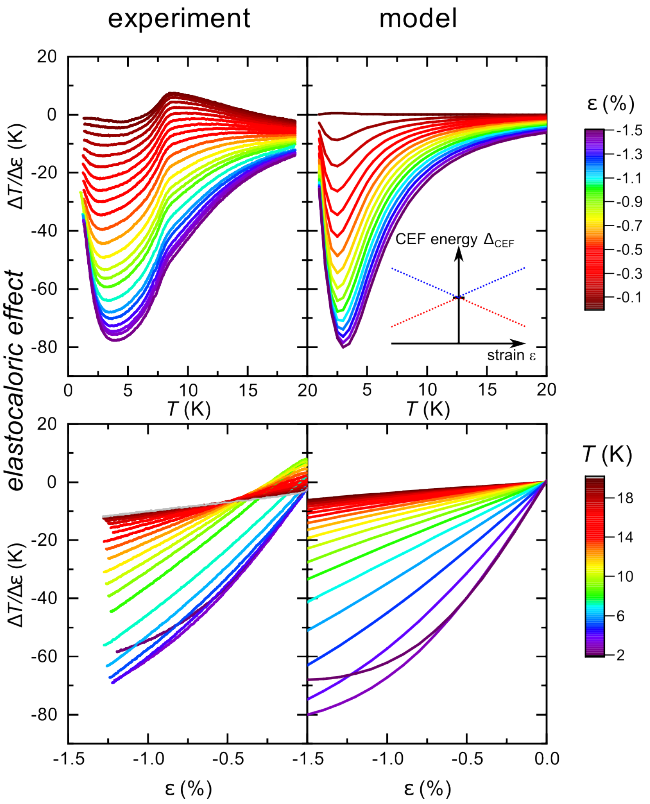

Fig. 1: Experimental data and theoretical modelling of the elastocaloric effect reveal signatures of the excited crystal-electric field levels in the 'super'-heavy fermion YbPtBi [3].

By explicitly breaking the high cubic symmetry by the application of uniaxial pressure, we identified clear signatures of a cubic crystal-electric field quartet with high degeneracy at low temperatures, which is split by strain (see Figure 1), [3]. By comparing the experimental values of the elastocaloric effect and theoretical modelling, we were able to determine on a quantitative level how these crystal-electric field levels change with strain. Thus, with our work, we not only shed light on the origin of the extremely high effective mass in this 'super'-heavy fermion, but also introduced a powerful approach for the strain engineering of crystal-field levels in quantum materials. This approach will be particularly relevant for the strain tuning of spin-orbit entangled frustrated magnets, where the crystal-electric field properties and parameters are crucial for the magnetic properties.

Spin-vortex magnetic phase stabilized by competing interactions

Another set of important results from this census period concerns the identification and tuning of the spin-vortex crystal (SVC) magnetic state. This type of order is considered exotic due to its chiral order parameter and its potential role as the low-entanglement parent state of the chiral spin liquid.

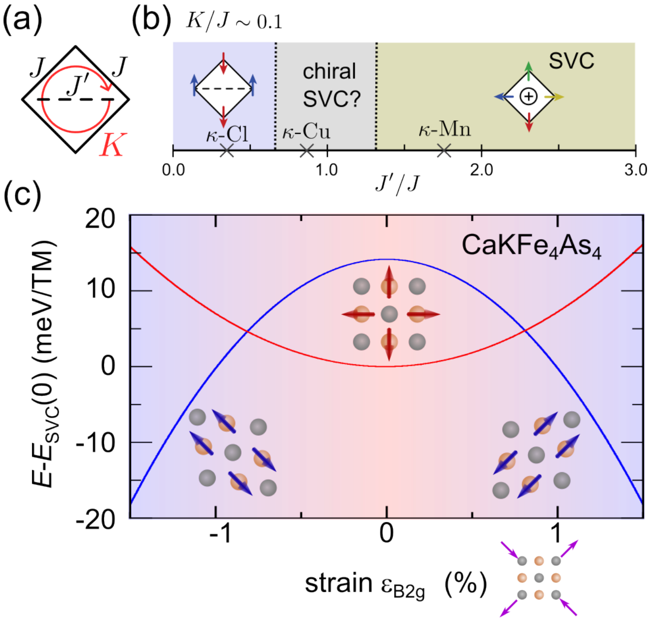

Fig. 2: The identification and strain control of the spin-vortex crystal (SVC) order with chiral order parameter; (a,b) In a frustrated Mott insulator, such as the organic charge transfer salt abbreviated with κ-Mn, the SVC is stabilized by exchange interactions J and J′ and a small ring exchange interaction K [5, 6]; (b) Moderate, experimentally accessible in-plane strains with B_{2g} symmetry can change the preferred SVC moment orientation to a stripe-antiferromagnetic one in the iron-based superconductor $CaKFe_4As_4$. [7].

Theoretical efforts in the recent years [4] have provided increasing evidence that the chiral spin liquid can be stabilized in a Heisenberg model on the triangular lattice with interactions J and J', when additional higher-order ring exchange interactions K are included (see Figure 2 a). The latter become highly relevant at intermediate correlation strengths in the proximity of the Mott metal-insulator transition.

The chiral spin liquid and its parent SVC state are predicted to exist only in a narrow range of intermediate correlation and large frustration, thus making it very challenging to stabilize these states experimentally. In a collaborative effort, involving thermodynamic and microscopic measurements and theoretical modeling, we successfully identified the SVC order in an organic triangular lattice compound [5, 6], whose chemical formula we abbreviate as κ-Mn (see Figure 2 a).

Next, we investigated the tunability of the SVC state by strain. Given the experimental challenge of developing protocols to strain fragile, organic crystals, we first focused our efforts on an inorganic, intermetallic material which shows the SVC order. Indeed, the iron-based superconductor $CaKFe_4As_4$ shows a tetragonal SVC ground state upon doping. It is worthwhile noting that this type of magnetic order is rare among the iron-based superconductors, which typically show a reduction of crystallographic symmetry when undergoing magnetic order. This motivated us to explore the impact of symmetry-breaking antisymmetric strains on the SVC state. We predicted that moderate strains within experimentally accessible ranges will induce a transition from the SVC state to a stripe-antiferromagnetic state, when the strain is applied along the in-plane [1 0 0] direction (i.e. a B_{2g} type strain) (see Figure 2 b). Experimentally, we were able to demonstrate that the properties of $CaKFe_4As_4$ are indeed strongly susceptible to this B_{2g} antisymmetric strain [7], but very robust against strains applied along other crystallographic directions. Our combined theoretical and experimental work concludes that antisymmetric strains are a powerful tuning parameter for unconventional magnetic states.

Preliminary work and future plans: Strain control of frustrated magnets

The achievements above – both in terms of methodology and physical understanding of strain effects on magnetic materials – form the foundation for our current and future research on the strain control of frustrated magnets. To highlight the potential, we present a specific preliminary example here. The organic triangular lattice magnet

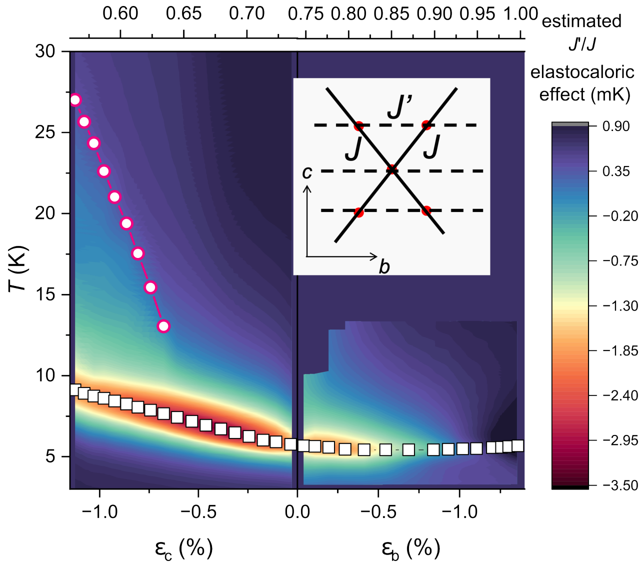

Fig. 3: Color map of the elastocaloric effect as a function of temperature and strain of a frustrated, organic triangular lattice magnet (compound abbreviated with κ-Cu). The experimental strain (bottom axis) adjusts the J'/J ratio (top axis), allowing a broad section of the phase diagram to be explored within a single experiment, including ordered states at low frustration and the ground state of the ideal isotropic triangular lattice.

κ-Cu (see Figure 2) possesses an almost isotropic triangular lattice at ambient pressure and is considered as a possible candidate for the realization of a gapped chiral spin-liquid ground state below $T^\star \sim 6\,\mathrm{K}$ [6]. By applying strain to κ-Cu to control the degree of frustration J'/J, we were able to vary J'/J by over 40%, allowing us to explore the properties of the triangular lattice in two opposite limits: (i) weak to moderate frustration and (ii) strong frustration of the isotropic triangular lattice, all within a single experiment (see Figure 3). By monitoring the elastocaloric effect, we can demonstrate the significant changes of the physical properties resulting from this substantial variation of J'/J. This is exemplified by the additional, possibly antiferromagnetic phase transition (open circles in Figure 3) that is observed at lower frustration ($J'/J \lesssim 0.65$). This data set will serve as an important experimental benchmark for theoretical calculations of the triangular-lattice Heisenberg model.

Method development: Dynamical properties of the lattice under AC strains

At least as interesting as the ground state properties of correlated and frustrated magnets are their dynamical properties. It is in this context that we aim to probe and tune the dynamics of correlated electron systems through our AC strain techniques. The conceptual idea, that we follow, is akin to the one of an AC susceptibility experiment, where the real and imaginary part of the magnetic susceptibility are determined by the application of an oscillating magnetic field. We have now adapted this idea to determine the "dynamic lattice susceptibility", or the AC Young's modulus. To determine Y_{AC}, we apply an AC stress and measure the induced AC strain. A finite imaginary part of Y_{AC} reveals information on the dynamical properties of the crystal lattice. It implies that there is a finite phase difference, δ, between the applied stress and the created strain, and the material is said to be viscoelastic, i.e., it adopts both solid-like and liquid-like properties.

While the study of viscoelastic properties through dynamic elastic measurements is well established in mechanical engineering, these have not yet been studied in the context of correlated quantum materials. In fact, the strategy to adopt methods from the field of mechanical engineering to ours has proven highly successful in the past. The recent work of this department on the nonlinear elasticity of Sr_2RuO_4 (see **PQM_04_Noad**) is an excellent example for this [2]. Through the implementation of stress-strain measurements, the work has yielded insights into the strong entanglement of electronic and lattice degrees of freedom in this correlated metal. The natural next step was to implement measurements of the dynamic stress-strain relationships in quantum materials, which we have achieved and described in Ref. [8].

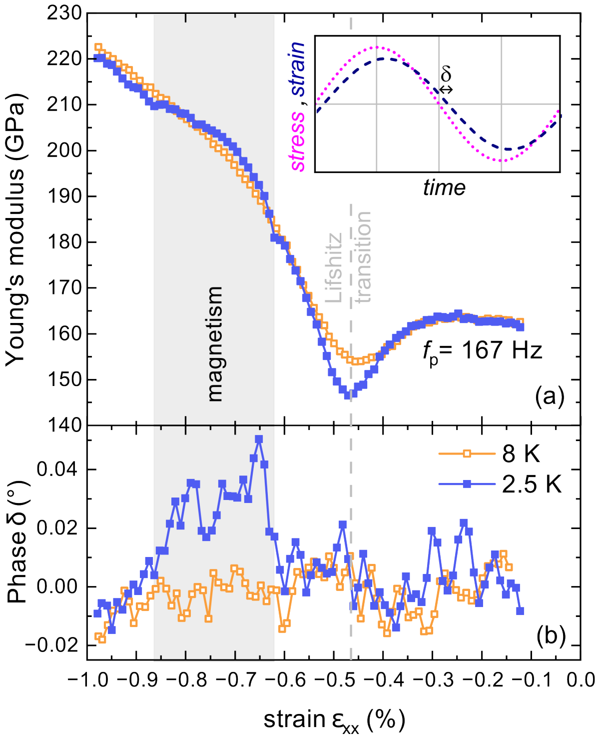

Fig. 4: Sr_2RuO_4 shows viscoelastic behavior, i.e. a finite phase difference between applied stress and resulting strain, when it orders magnetically at high compression and low temperatures.

We benchmarked our new experimental setup on Sr_2RuO_4 and results are shown in Figure 4. We were able to identify viscoelastic behavior in Sr_2RuO_4 when it is magnetic under high compression and at low temperatures. We are currently in the process of applying this technique to a wider range of stress-tunable magnetic quantum materials [9], e.g. to probe and to tune the intrinsic low-frequency dynamics of frustrated magnets by uniaxial pressure.

External Cooperation Partners

Our work on experimental strain tuning of correlated magnets benefits from collaborations with various groups nationally and internationally. We collaborate intensively with Paul Canfield (Ames National Laboratory/Iowa State University, Ames, USA); Roser Valentí, Michael Lang (Goethe Universität, Frankfurt); Takahiko Sasaki (Institute for Materials Research, Tohoku University, Sendai, Japan); Jochen Geck (TU Dresden); Stephen M. Winter (Wake Forest University, Winston-Salem, USA). We also actively participate in two Collaborative Research Centers, funded by the German Science Foundation, providing the formal framework for many of our collaborative activities: the Dresden-based CRC1143 on “Correlated Magnetism: From Frustration to Topology” as well as the CRC/TRR288 (Frankfurt-Mainz-Karlsruhe) “Elastic Tuning and Response of Electronic Quantum Phases of Matter”.

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