Stress-Strain Relation as a Probe of Quantum Materials

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In recent years, uniaxial pressure supplied by piezoelectric-based cells has become a powerful tuning parameter for studying quantum materials, but a key experimental challenge has been to combine it with thermodynamic measurements. We have developed a method for measuring the stress-strain relation under uniaxial pressure, allowing us to probe elastic moduli as a function of strain. Using this technique to study Sr_2RuO_4, we found a strong lattice softening at a Lifshitz transition. The simplicity of the experimental setup makes this new method compatible with a wide range of other probes, and the improved control over stress and strain gradients that was necessary for quantitative stress-strain will be beneficial for uniaxial pressure measurements more generally.

The piezoelectric-based uniaxial pressure cells pioneered by CWH in our department [1, 2] have made it possible to reliably tune quantum materials far from their ambient-pressure equilibrium crystal lattices. A major effort in PQM in recent years has been to develop methods for measuring thermodynamic quantities under uniaxial pressures. Such capabilities are highly desirable for probing the phase diagrams revealed with this tuning parameter but are challenging to achieve due to the necessarily strong linkages between sample and surrounding environment. In this report, I will describe one thermodynamic technique that has come to fruition over the past census period, involving measurements of the stress-strain relation. The work that I will describe [3] was the product of several years' effort which I (HMLN) undertook as a postdoc in the group led by CWH and am now carrying forward in my new role as a group leader.

Stress-strain as a thermodynamic probe

By measuring both the stress and resulting strain in a sample as we subject it to uniaxial pressure, we probe the stiffness of the material. From a thermodynamic perspective, this measurement probes strain derivatives of the free energy. Stress σ_{ij} is the first derivative of the free energy F with respect to strain ε_{ij}, while the elements of the stiffness tensor c_{ijkl} are the second derivative of the free energy with respect to strain:

$$c_{ijkl} = \frac{\partial^2 F}{\partial \varepsilon_{ij} \partial \varepsilon_{kl}}. \tag{1}$$

In uniaxial pressure measurements, in which the measured strain is along the same axis as the stress, a natural quantity to consider is Young's modulus, $E_\alpha = d\sigma_{\alpha\alpha}(\varepsilon_{kl})/d\varepsilon_{\alpha\alpha}$. As the strain derivative of stress, E_α probes second derivatives of the free energy with respect to strain; in our piezo-based cells, the sample is free to expand or contract in directions transverse to the applied pressure, and so the measured E_α will consist of several c_{ijkl} and the relevant Poisson ratios.

Stress-strain measurements at cryogenic temperatures

While ultrasound techniques [4] are well-established for probing c_{ijkl} in the limit of zero applied stress, few options exist for measuring elastic moduli at nonzero stresses at cryogenic temperatures. One approach, used with great success in [5] to demonstrate nonlinear elasticity around a critical endpoint in an organic compound, is to use a helium gas-based system to supply tunable hydrostatic pressures along with capacitive dilatometry for measuring strains. But at cryogenic temperatures, gas-based systems are best suited to relatively soft materials, because the maximum pressures they can supply are set by the solidification of the gas pressure medium.

Our approach was to build upon technological development within PQM from earlier census periods, using piezoelectric-driven uniaxial pressure cells that are designed to handle stiff and brittle millimeter-scale samples at cryogenic temperatures. In particular, we set out to use

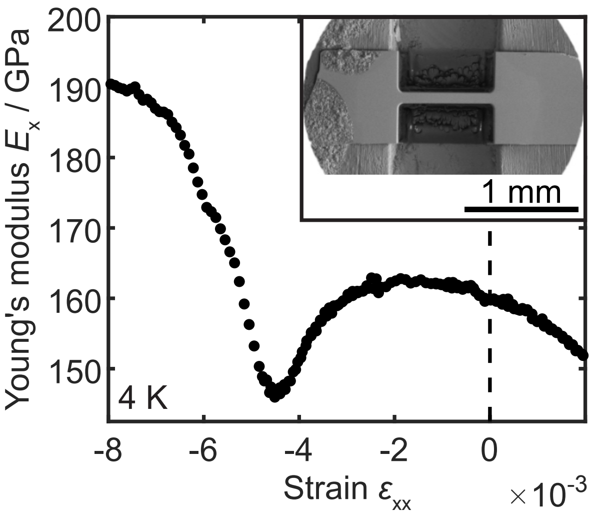

Fig. 1: Young's modulus as a function of strain in Sr_2RuO_4 under pressure applied along the x = [100] direction of the crystal at 4 K. Inset: SEM image of a sample after shaping with a focused ion beam.

a new variant with integrated capacitive force and displacement sensors [2]. Having access to both quantities simultaneously meant that we could check for undesired, irreversible deformations during an experiment, but also raised the possibility of finding intrinsically nonlinear elastic effects in samples driven through pressure-tuned phase transitions or regions of criticality.

For our first measurements, we turned to Sr_2RuO_4 because we were curious to see whether we might see any sign of the uniaxial-pressure-tuned Lifshitz transition [6, 7] in the stiffness of the lattice. More prosaically, the large body of existing work on Sr_2RuO_4, as well as the availability of centimeter-scale, high-purity single crystals [8] in PQM, made the material a useful testbed for developing and refining experimental methods and data analysis procedures.

Towards the end of the previous census period, we had made preliminary measurements of the stress-strain relation in Sr_2RuO_4 and could tell, at a qualitative level, that there were strong nonlinearities in the signal as a function of strain. To take full, quantitative advantage of the thermodynamic nature of the experiment, we worked out how to extract the response of the sample from the total measured signal. This involved developing procedures for shaping samples using a focused ion beam, as shown in Figure 1, to achieve well-defined regions of high and low stress in the sample. Since about half of the total measured displacement is taken up by the anchors, these additional steps were crucial for determining accurate values of stress and strain, and of the Young's modulus that we calculate from them.

Giant lattice softening at a uniaxial pressure-tuned Lifshitz transition

Our measurements of the stress-strain relation in Sr_2RuO_4 as a function of uniaxial pressure along $x = [100]$ revealed a large and strongly temperature-dependent softening of Young's modulus E_x at the Lifshitz strain (Figure 2a). Remarkably, this softening is driven by the conduction electron system. The temperature dependence tells us that it must be due to the entropy in the material, as opposed to the internal energy. At 4 K, the contribution of phonons to the entropy of Sr_2RuO_4 is negligible [9], as is the that of valence bands, which leaves only the conduction electron states to contribute. Indeed, as shown in Figure 2 of the Status Report **section 1.2**, the temperature-dependent softening in E_x is accompanied by a peak in the strain-dependent entropy.

A two-dimensional tight-binding model developed by our collaborators at the Karlsruhe Institute of Technology reproduces the softening in E_x (Figure 2b). All of the parameters in the model are constrained by prior experiments, and the strain dependence is included as a logarithmic contribution from the band that undergoes the pressure-tuned Lifshitz transition, reflecting the two-dimensionality of the electron system in Sr_2RuO_4.

In a material with three-dimensional band structure subjected to hydrostatic pressure, one would expect the relative softening of the lattice at a Lifshitz transition to be $\sim 10^{-4}$, three orders of magnitude smaller than the softening that we observe. In Sr_2RuO_4, we benefit from exceptionally clean crystals and a highly two-dimensional band structure, but the most important factor at our measurement temperatures is the sensitivity of the band structure to strain, parameterized by the deformation potential. The latter includes a factor α, which enters the magnitude of the softening as α^2. In our tight-binding model it is completely determined from prior experiments and has a value of 7.6, which, as it turns out, is close to the theoretically expected value of 8 for processes involving *d*-*p*-*d* orbital hopping, exactly as we have in the conduction electron system of Sr_2RuO_4.

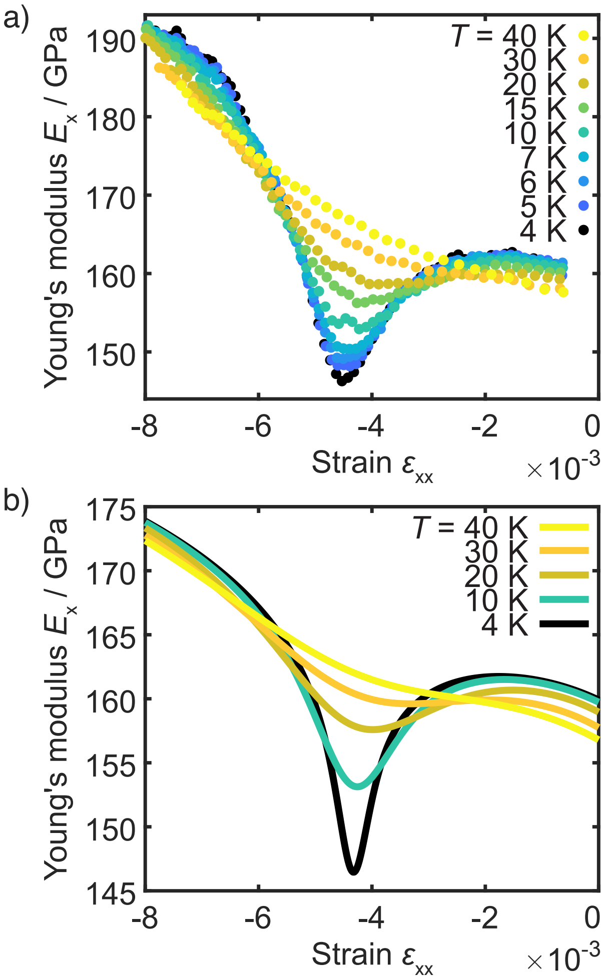

Fig. 2: Young's modulus of Sr_2RuO_4 as function of strain and temperature, (a) determined from our stress-strain experiments, (b) calculated in a tight-binding model.

Looking to the future

Our stress-strain measurements of Sr_2RuO_4 are just one example of the exciting physics that can be uncovered in thermodynamic measurements far from the ambient pressure axis. The piezo-cell-based stress-strain method is a highly adaptable tool; since it requires no wires on the sample, it is compatible with virtually any other probe that one might use in a uniaxial pressure measurement. Moreover, the FIB-based sample shaping that we developed for obtaining the Young's modulus in absolute units is more broadly useful, as it improves both the precision and accuracy of the strain in the experiment: the precision, through improving the homogeneity of stress and strain in the probed part of the sample, and the accuracy, by sharpening the definition of the strained length. In Figure 3, we show an example of such improvements in a measurement of the elastocaloric effect.

Furthermore, by reducing surface imperfections where cracks can nucleate, shaping samples with the FIB also helps us to achieve higher stresses in our experiments than with samples prepared by hand. This opens up new areas in phase space that we are excited to explore with our thermodynamic probes in coming years. To give just one example of what higher stresses can reveal, we recently found that the magnetic phase in Sr_2RuO_4, stabilized at compressions beyond the Lifshitz transition, exists only over a limited range of strains (see Figure 4a), and has a dome-shaped extent in temperature versus strain (Figure 4b).

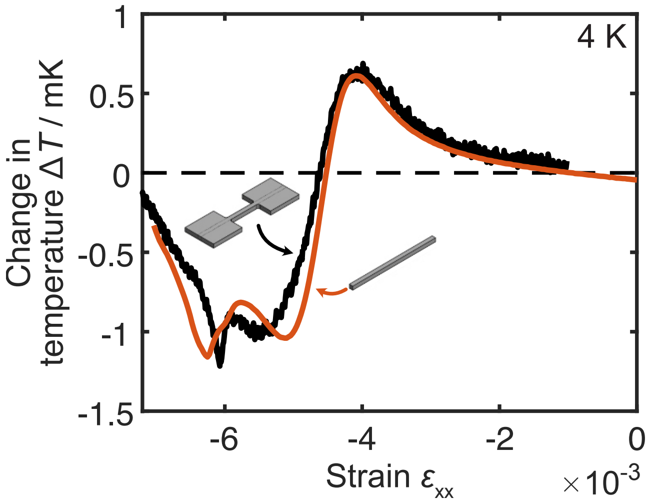

Fig. 3: Shaping samples for stress-strain measurements improves the strain resolution in other quantities measured at the same time. Features in the elasocaloric effect of Sr_2RuO_4 as a function of strain measured on a necked sample (black) are sharper than for a bar-shaped sample (orange, from Ref. [10]).

While the current generation of two-capacitor cells that we have used for the DC stress-strain technique are rather large, with a 4.6 cm outer diameter, and cannot be used for experiments requiring optical transmission through the sample, an AC version of the technique recently developed in PQM can be done in cells having only a displacement capacitor (see **PQM_03_Gati**) and Ref. [11] for more details). This opens the possibility of performing stress-strain measurements using existing variants of the uniaxial pressure cells that are smaller or designed for a transmission geometry.

Finally, measuring the stress-strain relation in an applied magnetic field requires no modification to the experimental setup since the sensing elements are capacitors, which are insensitive to magnetic field. We have begun using this additional tuning axis and look forward to having new results to share soon!

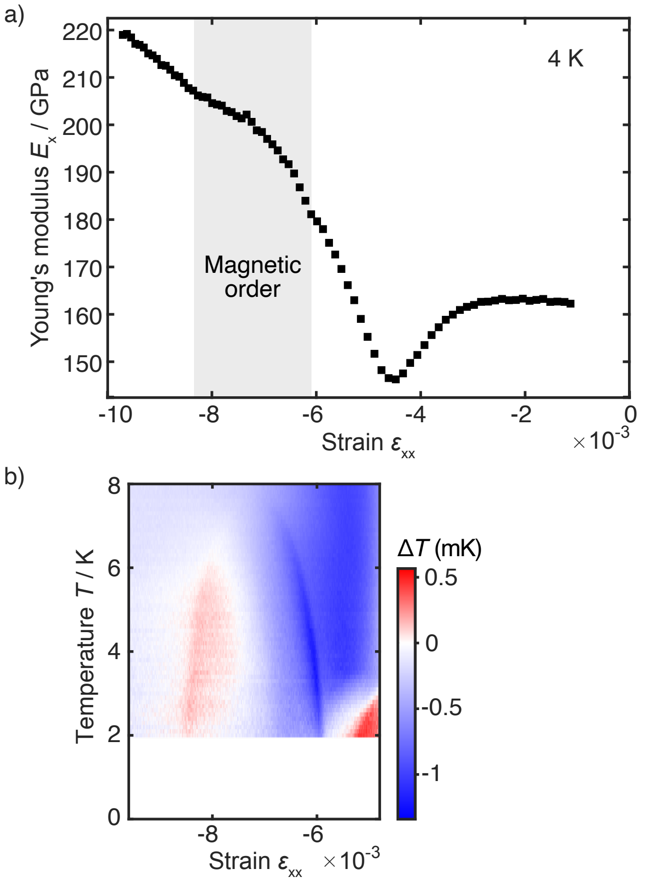

Fig. 4: Extending thermodynamic measurements of Sr_2RuO_4 to higher pressures along x = [100] reveals that a poorly understood magnetic phase is bounded in strain and forms a symmetric dome in temperature. (a) Young's modulus as a function of strain at 4 K and (b) preliminary temperature-strain map of the magnetic phase boundary from measurements of the elastocaloric effect on the same sample.

External Cooperation Partners

Our work on the stress-strain relation in Sr_2RuO_4 benefited greatly from collaborations with external partners both in Germany and abroad: Jörg Schmalian (Karlsruher Institute of Technology, Germany); Markus Garst (Karlsruher Institute of Technology, Germany); Igor Mazin (George Mason University, USA); Bongjae Kim (Kunsan National University, Korea); Naoki Kikugawa (National Institute for Materials Science, Japan). We would also like to acknowledge the broader collaborative environment of CRC/TRR288 (Frankfurt-Mainz-Karlsruhe) "Elastic Tuning and Response of Electronic Quantum Phases of Matter", funded by the German Science Foundation, in which HMLN, CWH, and APM all play an active role.

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