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Entropy landscape in strontium ruthenate

Elastocaloric determination of the phase diagram of Sr_2RuO_4
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Recommended with a Commentary by Qimiao Si, Rice University

Sr_2RuO_4 was for a long time considered to be a superconductor with odd-parity spin-triplet pairing, which is sought after for the possibility of hosting Majorana fermions. A new generation of NMR experiments [1, 2], however, have shown that the spin susceptibility in fact drops as temperature is lowered through the superconducting transition temperature. They have motivated considerable ongoing theoretical efforts to identify the pairing state for this venerable superconductor. The pairing functions are being constructed in the space of multiple orbitals, as has been extensively considered for iron-based superconductors, and candidate even-parity spin-singlet pairing states are being put forward (see, for example, Refs. [3, 4, 5]).

There are ample empirical reasons to consider that spin-singlet superconducting pairing develops near antiferromagnetic or related electronic orders. In the case of Sr_2RuO_4, it is unclear which part of the phase diagram hosts a long-range order of any kind other than superconductivity. One in principle fool-proof way to search for electronic orders or their fluctuations is to map out the entropy landscape. A genuine phase transition would be manifested through a singularity in the entropy (S) vs. temperature (T) or a non-thermal control parameter (pressure, strain, magnetic field, and so on). A quantum critical point (QCP) would be seen as a peak of the entropy as a function of the non-thermal control parameter [6, 7, 8, 9].

The highlighted study by Y.-S. Li and collaborators concerns entropy as a function of temperature and uniaxial strain in Sr_2RuO_4. To expand on how entropy vs. control parameter can be explored, consider the simplest case that the non-thermal control parameter is pressure p. A thermodynamic quantity of interest is thermal expansion $\alpha = (1/V)(\partial V/\partial T)_p$. Through a Maxwell's relation, it takes the following form,

$$\alpha \propto -\frac{\partial S}{\partial p}\,. \tag{1}$$

The ratio of α to the specific heat $c_p = (T/N)(\partial S/\partial T)_p$ defines the Grüneisen ratio, $\Gamma = \alpha/c_p$:

$$\Gamma \propto -\frac{\partial (S/T)/\partial p}{\partial S/\partial T}\,. \tag{2}$$

In the vicinity of a pressure-induced QCP, both p and T represent variables that are tuned on approach of the QCP ($T = 0$ and $p = p_c$) in the T-p phase diagram. It can then be seen from Eq. (2), via a dimensional analysis, that Γ manifests the response of the system to the control parameter p, which must be singular as the QCP is approached.

As a function of p in the low-T limit, this singularity has a particularly simple form. Dimensional analysis of Eq. (2) implies that Γ is inversely proportional to $p - p_c$. It follows from a scaling form for the free energy [8, 9] that the critical component Γ_{cr} is peaked at the QCP in a universal way,

$$\Gamma_{\mathrm{cr}} \propto \frac{1}{p - p_{\mathrm{c}}} . \tag{3}$$

In fact, even the scaling function cancels out in the proportionality factor. The predicted divergence of the Grüneisen ratio has been extensively verified in quantum critical heavy fermion metals [7, 10, 11]. Combining Eqs. (2,3) shows that entropy is maximized at the QCP.

When the tuning parameter is a magnetic field instead of pressure, the quantity of interest becomes the magnetocaloric effect [8]. Through the advent of iron-based superconductors, uniaxial strain has become increasingly widely used as a tuning parameter and, when it is considered in place of pressure, the quantity of interest is the elastocaloric effect. The method to measure the a.c. elastocaloric effect was recently developed in the context of iron-based superconductors [12], which have enhanced nematic correlations and are thus particularly responsive to uniaxial strain.

Y.-S. Li and collaborators carried out elastocaloric measurements for a substantial range of uniaxial strain (with a magnitude up to 0.7%) over the temperature range of interest to this system (between 1 K to 8 K). Not too surprisingly, entropy picks up the transition into the superconducting state.

What was unexpected is the evidence of another phase that develops with increasing strain. Entropy measurement by itself cannot specify the nature of this phase. However, muon spin relaxation measurement has indicated that the phase is antiferromagnetic [13]. The results encourage further magnetic measurements to firmly establish the existence of the phase and determine its nature.

A couple of remarks are in order. First, establishing an antiferromagnetic or related electronic order in proximity to superconductivity in the phase diagram of Sr_2RuO_4 would be significant. It would imply a degree of unification of this system with a variety of other correlated superconductors. Second, mapping out the entropic landscape is very informative about correlated systems with close-by phases and certainly deserves more frequent investigations in correlated materials. The entropic features reported here, while not especially pronounced, nonetheless illustrate the point.

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