Evidence for a nematic component to the hidden-order parameter in URu$_2$Si$_2$ from differential elastoresistance measurements

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For materials that harbour a continuous phase transition, the susceptibility of the material to various fields can be used to understand the nature of the fluctuating order and hence the nature of the ordered state. Here we use anisotropic biaxial strain to probe the nematic susceptibility of URu$_2$Si$_2$, a heavy fermion material for which the nature of the low temperature ‘hidden order’ state has defied comprehensive understanding for over 30 years. Our measurements reveal that the fluctuating order has a nematic component, confirming reports of twofold anisotropy in the broken symmetry state and strongly constraining theoretical models of the hidden-order phase.
The heavy fermion compound URu$_2$Si$_2$ undergoes a continuous phase transition at $T_{HO} \sim 17$ K, the precise nature of which has not been fully established, despite considerable experimental and theoretical attention. Determination of the symmetry (or symmetries) that are broken in the ‘hidden order’ phase is not just of purely academic interest, but also has a direct bearing on the symmetry of the superconducting gap that develops below $T_c = 1.5$ K. Of particular relevance to the present work, several recent measurements have provided evidence that the hidden-order phase breaks the fourfold rotational symmetry of the high-temperature tetragonal crystal lattice, however, the degree of anisotropy that is observed depends heavily on the crystal size (as determined via torque measurements) and also on the crystal quality (for X-ray diffraction measurements), leading to some contention as to whether or not these are intrinsic effects. In the current work, we take a complementary approach and probe the fluctuating order in the temperature regime above $T_{HO}$. Our results demonstrate first that the fluctuating order has a nematic component, confirming that the hidden-order phase spontaneously breaks fourfold rotational symmetry, and second that other symmetries must also be broken at the phase transition.

To measure a general susceptibility of a system, one must apply the appropriate conjugate field (that is, the physical quantity that couples linearly to the given order) and measure the linear response. For a tetragonal material, such as URu$_2$Si$_2$ above the hidden-order transition, the conjugate field to electronic nematic order is anisotropic biaxial in-plane strain (either $\varepsilon_{aniso} = (\varepsilon_{xx} - \varepsilon_{yy})$ or $\varepsilon_{aniso} = 2\varepsilon_{yy}$ for spontaneous order in the [100] or [110] directions, corresponding to $B_{1g}$ and $B_{2g}$ representations of the $D_{4h}$ point group, respectively). The nematic susceptibility for each of these orientations is then $\chi_N \propto \lim_{\varepsilon_{aniso} \to 0} \frac{\partial \rho}{\partial \varepsilon_{aniso}}$, where $\rho$ represents any thermodynamic quantity measuring the induced anisotropy in mutually orthogonal directions at the phase transition, signalling an instability towards nematic order. Such behaviour was recently observed for the representative underdoped iron pnictide Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$, demonstrating that the structural phase transition that precedes the onset of collinear antiferromagnetic order in that material is driven by electronic nematic order.

There are also classes of order for which there is a single transition to an ordered state that has a nematic component, while simultaneously breaking additional symmetries. A simple example would be a unidirectional incommensurate density wave with order parameter $\Delta = \Delta(Q)$, for which both translational and tetragonal symmetries are simultaneously broken at the same transition (Q corresponds to the ordering wave vector of the density wave, and $i = x$ or $y$). In such a case, anisotropic strain is not conjugate to the order parameter (that is, the coupling term in the free-energy expansion is not bilinear). Nevertheless, the inherent nematicity associated with the unidirectional density wave motivates introduction of a nematic order parameter $\mathcal{N}$, which couples linearly to $(\Delta_{x}^2 - \Delta_{y}^2)$ and to which anisotropic strain is linearly coupled (see ‘Nematic × (?) Symmetry’ subsection in the Supplementary Discussion). For such a situation, and with the assumption of a continuous transition, an analysis based on standard Ginzburg–Landau theory for coupled order parameters reveals that the nematic susceptibility follows a Curie–Weiss temperature dependence at high temperatures as the density wave fluctuations orient in the anisotropic strain field, but then develops an additional divergence close to the transition temperature, experimentally distinguishing the behaviour from that of a ‘pure’ nematic transition. We find that hidden order in URu$_2$Si$_2$ appears to fall into this latter class, having a nematic component but also breaking additional symmetries at the hidden-order phase transition (see ‘Nematic Susceptibility: Theory’ subsection in the Supplementary Discussion).

Our measurements are based on a novel technique that probes the differential response in the electrical resistivity to anisotropic biaxial strain in mutually perpendicular directions. We measure the induced resistivity anisotropy, $N = \frac{\rho_{xx} - \rho_{yy}}{\rho_{xx} + \rho_{yy}} \sim \left( \frac{\Delta R/R_{yy}}{\Delta R/R_{xx}} \right)$, which, by symmetry, is proportional to $\psi$ in the limit of asymptotically small values. The induced anisotropy is characterized by specific terms in the associated elastoresistivity tensor $\mathcal{M}$ that relate changes in the resistivity to strains experienced by the material. In the regime of infinitesimal strains (linear response), the elastoresistivity coefficients $(m_{11} - m_{12})$ and $2m_{66}$ are linearly proportional to the bare (unrenormalized) nematic susceptibility, $2\chi_{N,0}$ and $\chi_{N,0}$, for strains in the [100] and [110] directions, respectively. The proportionality constant relating the resistivity anisotropy to the nematic order parameter is governed by microscopic physics, but does not contain any singular contributions. Consequently, any divergence of the induced resistivity anisotropy is directly related to divergence of the nematic susceptibility. Since this is ultimately a derivative of a transport measurement, the technique is especially sensitive to static or fluctuating order that affects the Fermi surface.

As we describe in this study, our measurements reveal a strongly anisotropic differential elastoresistance between the $B_{1g}$ and $B_{2g}$ symmetry channels. In the $B_{2g}$ channel for temperatures well above $T_{HO}$, the $2m_{66}$ elastoresistivity coefficient follows a Curie–Weiss dependence. This behaviour directly implies that the fluctuating order has a nematic component. However, we also find a striking additional divergence in $2m_{66}$ close to the phase transition, which tracks the heat capacity. This additional contribution to the differential elastoresistance demonstrates that while hidden order in URu$_2$Si$_2$ must break fourfold rotational symmetry, it also breaks other symmetries and hence is not purely nematic.

**Results**

Representative measurements of $(\Delta R/R_{yy})$ for five different temperatures are shown in Fig. 1b,c for [100] and [100] oriented crystals, respectively, revealing a linear response. The sign of $(\Delta R/R_{xx})$ is opposite to that of $(\Delta R/R_{yy})$ due to the anisotropic strain (that is, the imposed symmetry breaking), distinct from changes in resistance observed under conditions of hydrostatic pressure.

This procedure was performed for both the $(\Delta R/R_{xx})$ and $(\Delta R/R_{yy})$ orientations on the same sample by removing the crystal from the piezo with acetone between each measurement. The slopes $\Delta R/R$ versus strain for each orientation were found using a linear fit and the difference was taken. The resulting elastoresistivity coefficients $2m_{66}$ and $m_{11} - m_{12}$ are plotted in Fig. 2 as a function of temperature.

The data shown in Figs 1 and 2 reveal a striking anisotropy in the elastoresistivity coefficients, with $2m_{66}$ values considerably larger, and also more strongly temperature dependent, than $m_{11} - m_{12}$. For comparison, the elastoresistivity coefficients of simple metals are small (of order one) and essentially isotropic.

It is worth remarking that the differential elastoresistance of URu$_2$Si$_2$ is comparable to that of BaFe$_2$As$_2$, for which the response along the [100] and [110] orientations is also strongly anisotropic, with $2m_{66}$ values reaching ~40 approaching the tetragonal-to-orthorhombic phase transition in that material.
Figure 1 | Measurement of elastoresistivity coefficients. Anisotropic strain is achieved by gluing thin crystals of URu2Si2 to the side of a PZT piezoelectric stack. Strain gauges mounted on the opposite face of the piezo stack measure the range of temperatures over which this fit can be applied is elastoresistivity only develops significant values below B. In contrast, in the [110] direction the temperature derivative of the in-plane resistivity dT for [100] orientation due to imperfectly constrained current paths within the crystal. For the remaining discussion we focus solely on the [100] direction.

Recalling that 2m66 is proportional to the nematic susceptibility in the [110] direction (ξN110), the large values of 2m66 (red data points in Fig. 2) and the strong temperature dependence both indicate a diverging nematic susceptibility in the B2g (dxy symmetry) channel, and hence motivate fitting the data to the Curie–Weiss form (2m66 = C/[(T - θ)] + 2m66). Since the elastoresistivity only develops significant values below ~60 K, the range of temperatures over which this fit can be applied is necessarily small. Nevertheless, if we attempt fits over different temperature ranges, the best fit, defined by a reduced χ2 statistic closest to 1, occurs for a range from ~30 K to the maximum of 60 K, with a Weiss temperature θ = 15.2 ± 0.3 K (see ‘Curie Fitting’ subsection in the Supplementary Discussion). Extrapolation of the best fit function to lower temperatures (shown as a green line in Fig. 2) reveals significant deviation from Curie–Weiss-like behaviour for temperatures below ~30 K. The onset of this deviation at 30 K is coincident with a suppression in (T1T)-1 seen in recent NMR experiments21, suggestive of a common origin, possibly associated with the progressive development of strongly fluctuating order.

In contrast, m11 – m12, which is proportional to ξN100, is small and exhibits only a weak temperature dependence (cyan data points in Fig. 2); additional measurements on a separate crystal confirmed the small value of m11 – m12 (see ‘Elastoresistivity Measurements’ subsection in the Supplementary Discussion). Given the large values of 2m66, it is likely that this weak temperature dependence derives from slight sample misalignment (that is, a small amount of contamination from 2m66 in the [100] orientation due to imperfectly constrained current paths within the crystal). For the remaining discussion we focus solely on the 2m66 data.

The most striking aspect of the temperature dependence of the elastoresistivity coefficients is the sharp downward anomaly observed at THO. The 2m66 data are replotted in Fig. 3a on an expanded scale for the temperature window from 15.5 to 19 K, spanning THO. The anomaly exactly aligns with the sharp peak in the temperature derivative of the in-plane resistivity dp/dT (Fig. 3b,c), indicating that the effect is associated with the thermodynamic phase transition at THO. For comparison, heat-capacity measurements were also performed for the same crystal that was used for the [110] oriented measurements. The data closely follow the temperature dependence of dp/dT (ref. 22), reminiscent of the theoretical treatment of critical fluctuations in metallic antiferromagnets by Fisher and Langer23, with slight differences in the peak position attributed to small differences in
thermometry between the two different systems used for transport and thermodynamic measurements.

**Discussion**

The observation of the Curie–Weiss temperature dependence of the $2m_{66}$ elastoresistivity coefficient, combined with the large anisotropy between $2m_{66}$ and $m_{11} - m_{12}$, strongly implies a nematic character to the fluctuations associated with the hidden-order phase. However, the sharp downward anomaly in resistivity near the hidden-order transition, which is proportional to the square of hidden order that generates a nematic distortion; within mean field considerations, both nematic and energy density fluctuations (that is, the heat capacity) are related to the square of the order parameter.

As described previously, the nematic susceptibility of a material that suffers an electronic nematic instability follows a Curie–Weiss temperature dependence, as has been observed for the iron pnictides. However, a nematic response is also possible if the ordered state breaks fourfold spatial rotation symmetry, but does not couple to strain as a bilinear term in the free energy. In this scenario, the hidden order state would be described by a two-component vector-order parameter $\mathbf{\Delta} = (\Delta_x, \Delta_y)$. Motivated by the inherent nematicity in such a system, the nematic response can be modelled in mean field theory by introducing a separate nematic order parameter $\mathcal{N}$, which couples to the strain in the form

$$\Delta F = \epsilon_{\text{aniso}} \mathcal{N} + \lambda \mathcal{N}^2 \left( |\Delta_x|^2 - |\Delta_y|^2 \right),$$

where $\lambda$ is a coupling constant. The theory does not depend on the microscopic origin of $\mathcal{N}$, which could be an independent-order parameter or parasitic to the hidden order. Within such a theory, an additional contribution to the nematic susceptibility occurs close to the hidden-order transition, which is proportional to the singular part of the heat capacity ($\Delta C_V$) associated with the critical behaviour:

$$\chi_{\mathcal{N}} \sim \frac{1}{T - T_{\text{HO}}} (1 + \beta \Delta C_V),$$

where the constant $\beta$ depends on the strengths of the couplings between $\mathcal{N}$, $\mathbf{\Delta}$, and $\epsilon_{\text{aniso}}$ (see ‘Nematic × (?) Symmetry: Functional Form’ subsection in the Supplementary Discussion). This effect is analogous to anomalies observed in the elastic moduli and thermal expansion coefficients at a structural phase transition, first described by Testardi. Intuitively, the relationship between the nematic susceptibility and heat capacity arises because it is the square of hidden order that generates a nematic distortion; within mean field considerations, both nematic and energy density fluctuations (that is, the heat capacity) are related to the square of the order parameter.
The close correspondence between the predictions of this Ginzburg–Landau treatment and the observed data strongly suggest that hidden order in URu$_2$Si$_2$ is described by a two-component order parameter that breaks fourfold symmetry. Since the $2m_{66}$ coefficient diverges but $m_{11} - m_{12}$ does not, the orientation of the nematic fluctuations is along the [110] and [110] directions, consistent with the orientation deduced by measurements in the broken symmetry state$^{13}$.

The significance of this result is not so much the deduction that other symmetries are broken in the hidden-order phase. Over a period of many years, a growing body of evidence points to partial gapping of the Fermi surface and a possible doubling of the unit cell in the $c$-direction$^{34–36}$. Rather, our result is significant because it confirms the more controversial conclusions drawn from earlier torque$^{13}$, NMR$^{15}$, cyclotron resonance$^{16}$ and X-ray diffraction$^{14}$ measurements that the hidden-order phase spontaneously breaks fourfold lattice symmetry. Our results, which are ultimately based on statements about symmetry, do not identify a specific microscopic model for hidden order, but they clearly place tight constraints, namely that the hidden-order phase is described by a vector-order parameter that breaks fourfold symmetry.

The same coupling to the crystal lattice that yields the resistive response to strain must also cause an orthorhombic distortion below $T_{HO}$, consistent with reports of high-resolution X-ray diffraction measurements$^{14}$. Whether or not this anisotropy is observed in thermodynamic measurements of macroscopic crystals is then a matter of the resolution of the specific measurement and the relative population of the two orthogonal orthorhombic domain orientations (which will be influenced by both crystal size and quality, via effects associated with pinning of domain walls). Both aspects illustrate the distinct advantage of probing the nematic susceptibility in the temperature regime $T > T_{HO}$, for which there are no domains (one is simply probing the susceptibility of the symmetric phase) and for which the resistivity anisotropy is extremely sensitive to subtle perturbations of the Fermi surface.

Despite these advances, important questions remain. In particular, since nematic fluctuations in the $B_{2g}$ channel couple linearly to the shear modulus, one anticipates a softening of the $C_{66}$ elastic constant approaching the hidden-order phase transition. However, earlier measurements of the elastic moduli$^{37,38}$ do not show such a softening. At least in principle, weak coupling between the nematic-order parameter and the crystal lattice (consistent with the small magnitude of the orthorhombic distortion observed in X-ray diffraction experiments$^{14}$) would limit any appreciable renormalization of $C_{66}$ to small values of the reduced temperature close to the phase transition (see ‘Nematic Susceptibility: Theory’ subsection in the Supplementary Discussion). Nevertheless, the present results, in conjunction with other measurements that reveal a twofold anisotropy in the hidden-order phase$^{13}$, clearly motivate a careful reinvestigation of the elastic properties of URu$_2$Si$_2$ for samples of a comparably high RRR.

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Author contributions
E.D.B. and R.E.B. grew the crystals. S.C.R., M.C.S. and I.R.F. conceived of the experiment. M.W. and I.R.F. performed the differential elastoresistance measurements. A.V.M. and P.G.G. performed additional X-ray diffraction measurements on the crystals. A.V.M. and S.R. contributed to the theory. M.C.S., A.V.M., S.R., E.D.B., R.E.B. and I.R.F. contributed to writing the paper.

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