# Electronic Nematicity in $URu_2Si_2$ Revisited

Liran Wang,[1,*] Mingquan He,[1,*] Frédéric Hardy,[1] Dai Aoki,[2,3] Kristin Willa,[1] Jacques Flouquet,[2] and Christoph Meingast[1,†]
[1]*Institute for Quantum Materials and Technologies, Karlsruhe Institute of Technology, 76021 Karlsruhe, Germany*
[2]*Université Grenoble Alpes, CEA, PHELIQS, 38000 Grenoble, France*
[3]*Institute for Materials Research, Tohoku University, Oarai, Ibaraki 311-1313, Japan*

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The nature of the hidden-order (HO) state in $URu_2Si_2$ remains one of the major unsolved issues in heavy-fermion physics. Recently, torque magnetometry, x-ray diffraction, and elastoresistivity data have suggested that the HO phase transition at $T_{HO} \approx 17.5$ K is driven by electronic nematic effects. Here, we search for thermodynamic signatures of this purported structural instability using anisotropic thermal expansion, Young's modulus, elastoresistivity, and specific-heat measurements. In contrast to the published results, we find no evidence of a rotational symmetry breaking in any of our data. Interestingly, our elastoresistivity measurements, which are in full agreement with published results, exhibit a Curie-Weiss divergence, which we however attribute to a volume and not to a symmetry-breaking effect. Finally, clear evidence for thermal fluctuations is observed in our heat-capacity data, from which we estimate the HO correlation length.

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Despite 35 years of intensive experimental and theoretical efforts [1,2], the microscopic nature of the hidden-order (HO) state in $URu_2Si_2$ is unknown and remains one of the major unsolved issues in heavy-fermion physics. Recently, torque magnetometry, x-ray diffraction, and elastoresistivity have reported experimental signatures of electronic nematicity at the hidden-order phase transition $T_{HO} \approx$ 17.5 K, which, when confirmed, would narrow down the possible order parameters [3–5]. These reports point to a crystallographic symmetry lowering at the HO transition from a tetragonal to an orthorhombic structure, which would favor several theories, e.g., those involving multipolar orders, which rely on the breaking of the fourfold symmetry below $T_{HO}$ (see Ref. [6]). However, another recent x-ray diffraction study found no evidence of a structural transition [7], and improved nuclear-magnetic resonance (NMR) experiments now suggest an odd-parity electronic multipolar ordering within a tetragonal environment [8], although previous NMR data pointed to a twofold ordering [9].

In this Letter, in order to resolve the above controversy, we use three different sensitive experimental techniques to search for bulk experimental evidence of the purported nematic order parameter in well-characterized $URu_2Si_2$ single crystals [10]. First, we utilize anisotropic high-resolution capacitance dilatometry, which is several orders of magnitude more sensitive than the x-ray diffraction measurements reported in Ref. [4], and has recently been used to study nematicity in Fe-based systems [11–13]. Second, a symmetry-breaking transition inevitably leads to a drastic softening of its associated shear modulus, and we have thus performed Young's modulus measurements using a three-point-bending setup, which has been shown to be a very sensitive technique for detecting lattice softening in Fe-based materials [11,12,14,15]. Finally, we study the elastoresistivity [16] as a third sensitive method for observing nematicity. Our main result is that we find absolutely no evidence for a symmetry-breaking transition in either the thermal expansion or the Young's modulus measurements. Our elastoresistivity data interestingly exhibit a Curie-Weiss divergence, similar to the results of Riggs *et al.* [5]. However, we find that a very similar behavior can be inferred from hydrostatic-pressure measurements, revealing that a Curie-Weiss-like response under an enforced symmetry-breaking strain does not necessarily imply a nematic origin.

Single crystals of $URu_2Si_2$ were prepared by the Czochralski method and annealed at high temperature under ultrahigh vacuum, as described in detail in Refs. [17,18]. The residual resistivity ratio is typically around 100, indicating high-quality crystals. Thermal-expansion measurements were carried out on a single crystal ($2.0 \times 1.8 \times 2.0$ $mm^3$) using a homebuilt high-resolution capacitance dilatometer [19]. Heat-capacity measurements were made on the same crystal with the physical properties measurement system (PPMS) from Quantum Design using the dual-slope method [20,21]. Young's modulus data were obtained with the same dilatometer set up in a three-point-bending configuration [14,15]. A sketch of this setup, in which the force from the dilatometer springs causes a deflection of the crystal, is shown in Fig. 3. Elastoresistivity measurements were made by gluing a crystal on a glass-fiber-reinforced plastic substrate as described in Ref. [22] and in the Supplemental Material [23].

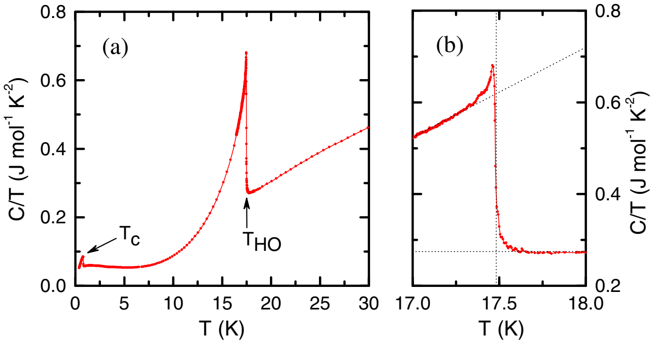

FIG. 1. (a) Heat capacity of our $URu_2Si_2$ single crystal. The data around $T_{HO} = 17.5$ K are plotted on an enlarged scale (b). The high quality of our single crystal is attested by the sharpness of the transition, allowing thermal fluctuations of the HO order parameter to be clearly observed.

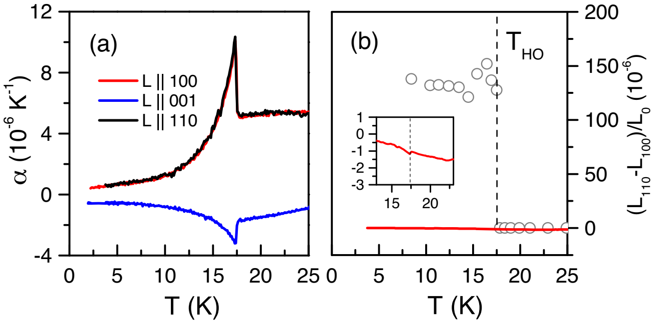

FIG. 2. (a) Thermal-expansion coefficients along [100], [001], and [110] directions near the hidden-order transition. (b) Orthorhombic distortion, i.e., the difference in thermal expansion along [100] and [110] compared to the x-ray diffraction data of Tonegawa *et al.* [4] (open circles). No symmetry-breaking distortion can be observed in our data (solid red line).

The HO transition results in a prominent anomaly at $T_{HO} = 17.49$ K in the heat capacity as illustrated in Fig. 1(a). Clear signs of order-parameter fluctuations in a range of $\approx 0.1$ K above and below $T_{HO}$ [see Fig. 1(b)] are observed, from which we estimate a Ginzburg parameter of $G_i = 0.006$. To the best of our knowledge, this is the first time that such fluctuations have been observed, and from $G_i$ and the "condensation energy" $H_c = 0.47$ T (derived from specific heat), we estimate a correlation length of 1 nm using standard Gaussian fluctuation theory. This short correlation length is an indication of a very short-range interaction leading to the HO state. From the rounding of the anomaly, we estimate that the transition width is as small as 0.015 K, clearly demonstrating the high homogeneity of our crystals. Systematic studies on different crystals have shown that the HO phase is very robust, i.e., not strongly impurity dependent [18].

A large $C_4$-symmetry-breaking strain, as reported by x-ray diffraction [4] below $T_{HO}$, should be easily detected using our dilatometer by comparing the strains $\varepsilon_{100}(T) = (\Delta L_{100}/L_{100})$ and $\varepsilon_{110}(T) = (\Delta L_{110}/L_{110})$ measured along [100] and [110] directions, respectively, as has been demonstrated for Fe-based materials [26–28]. This is because our spring-loaded dilatometer exerts a non-negligible stress along the measurement direction, and thus, for a measurement along the tetragonal [110] direction, the population of possible structural domains (twins) with the shorter orthorhombic axis should be favored by this stress. This would result in an *in situ* detwinning of the sample below $T_{HO}$, if the crystal symmetry were lowered. On the other hand, the twin population would remain unaffected by the dilatometer force for measurements along the [100] direction, which probe a mixture of both orthorhombic axes. We note that for the present measurements we have used "hard" springs, which apply a large force of about 300 g (3 MPa) to the crystal, i.e., about a factor of 5 larger than in our previous experiments on pnictides [26–28]. As illustrated in Fig. 2(a), we find no measurable difference between the coefficients of linear thermal expansion $\alpha_{100}(T) = (\partial\varepsilon_{xx}/\partial T)$ and $\alpha_{110}(T) = (\partial\varepsilon_{xy}/\partial T)$ measured along the [100] and [110] directions, respectively. To quantitatively compare our data with the x-ray diffraction results [4], we plot in Fig. 2(b) the orthorhombic distortion, i.e., the normalized difference in length $(L_{110} - L_{100})/L_0$. Clearly, our data, which show no signature of any distortion (see red line), are incompatible with the large distortion reported in Ref. [4]. Our resolution limit is about 700 times smaller than the reported distortion [see inset in Fig. 2(b)] [23]. We note that our results are in excellent agreement with those of de Visser and co-workers [29,30] published more than two decades ago. Additionally, the diffraction-inferred strain changes discontinuously at $T_{HO}$, which seems at odds with the clear second-order nature of the transition, observed in both specific-heat and thermal-expansion measurements; see Figs. 1 and 2(a).

Another powerful method for searching for a nematic instability is to measure the relevant shear modulus ($c_{66}$ elastic constant for $URu_2Si_2$), which necessarily has to approach zero as the transition is approached from above. As shown previously for $BaFe_2As_2$, the appropriate Young's modulus is in a good approximation proportional to $c_{66}$ [11,12,14,15]. For $URu_2Si_2$ the relevant Young's moduli, $Y_{100}$ and $Y_{110}$, can be determined using the three-point-bending technique with the tetragonal [100] and [110] crystal axis perpendicular to the beam supports, respectively (see sketch in Fig. 3). Here the moduli are expressed in terms of the elastic constants $c_{ij}$ by

$$Y_{100} = (c_{11} - c_{12})(1 + \eta)$$

and

$$Y_{110} = \left(\frac{1}{c_{66}} + \frac{1}{\gamma}\right)^{-1},$$

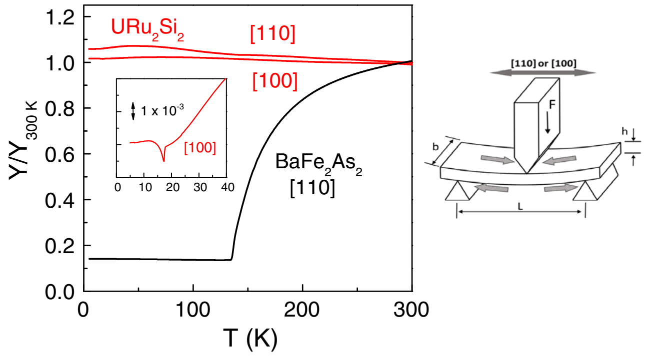

FIG. 3. Young's moduli of $URu_2Si_2$ measured along the [100] and [110] directions using a three-point-bending setup (see right-hand sketch). In contrast to $BaFe_2As_2$, we find absolutely no evidence for any soft mode in $URu_2Si_2$. The left-hand inset shows a magnified view of the [100] data around $T_{HO}$.

with $\eta = (c_{12}c_{33} - c_{13}^2)/(c_{11}c_{33} - c_{13}^2)$ and $\gamma = (c_{11} + c_{12})/2 - c_{13}^2/c_{33}$. Near a structural transition the shear mode will soften significantly, with $c_{66} \ll \gamma$ and, thus,

$$Y_{110} \propto c_{66}.$$

In Fig. 3, we compare $Y_{100}$ and $Y_{110}$ of $URu_2Si_2$ to the soft shear mode of $BaFe_2As_2$ ($Y_{110}$). In contrast to the strong softening observed in $BaFe_2As_2$ at the spin-density-wave transition $T_{s,N} = 140$ K, both Young's moduli of $URu_2Si_2$ increase upon cooling, as expected for phonon hardening, and exhibit absolutely no evidence for any kind of soft-mode behavior. In Ref. [4] it was argued that the antiferro nature of HO makes the elastic constant ($C_{11}$-$C_{12}$), being sensitive to $Q = 0$, to only couple weakly to the proposed symmetry breaking. Even for weak coupling, one would nevertheless expect a drastic softening of the relevant elastic mode, however in a smaller temperature interval.

We note that the top (bottom) surface of the bar-shaped sample experiences even larger compressive (tensile) stresses (≈15 MPa) in these bending configurations than in our thermal-expansion measurements [12], and, because we see no softening even under these extreme conditions, we conclude that there exists no evidence for a $C_4$-symmetry reduction in $URu_2Si_2$ even under the quite large uniaxial strains of 3–15 MPa. As seen in Fig. 3, both moduli do soften very slightly below approximately 70 K, as reported earlier for $(c_{11} - c_{12})/2$ by ultrasound investigations, which, however, is not indicative of a soft mode, but rather indicates a coupling of the lattice to the quadrupolar moment as argued by Kuwahara *et al.* [30]. Additionally, there is the small expected softening at $T_{HO}$ resulting from the pressure or strain dependence of $T_{HO}$ (see inset of Fig. 3).

Finally, we examine the elastoresistivity $2m_{66} = (\rho_{x'x'} - \rho_{y'y'})/[(1/2)(\rho_{x'x'} + \rho_{y'y'})(\epsilon_{x'x'} - \epsilon_{y'y'})]$ [see Fig. 4(a) and Ref. [23] for the definition of the coordinates], which has been argued to be a measure of the nematic susceptibility in Fe-based superconductors [16]. In Fig. 4(a) we present our result of $2m_{66}$ determined using a differential thermal-expansion method [22]. Here, the crystal was glued to a fiber-glass substrate, which exhibits significant anisotropic thermal expansion, so that the crystal experiences an anisotropic strain upon cooling. The strain transmitted to the crystal is estimated by measuring the thermal expansion of the crystal and substrate separately [22,23]. Using the same four electrical contacts, the resistivity difference between the crystal in freestanding and strained configurations provides the anisotropic strain response.

A sizable strain dependence of the resistivity is observed, with very similar $2m_{66}$ values and temperature dependence, as determined previously using a piezostack by Riggs *et al.* [5]. In particular, our data above about 30 K can also be fit with a Curie-Weiss temperature dependence, $2m_{66} = C/(T - T_\theta) + 2m_{66}^0$, with $T_\theta = 16$ K, which is slightly lower than $T_{HO} = 17.5$ K, suggestive of a sizable nematic response. However, as argued below, this suggested nematic response in elastoresistivity, which is at odds with our

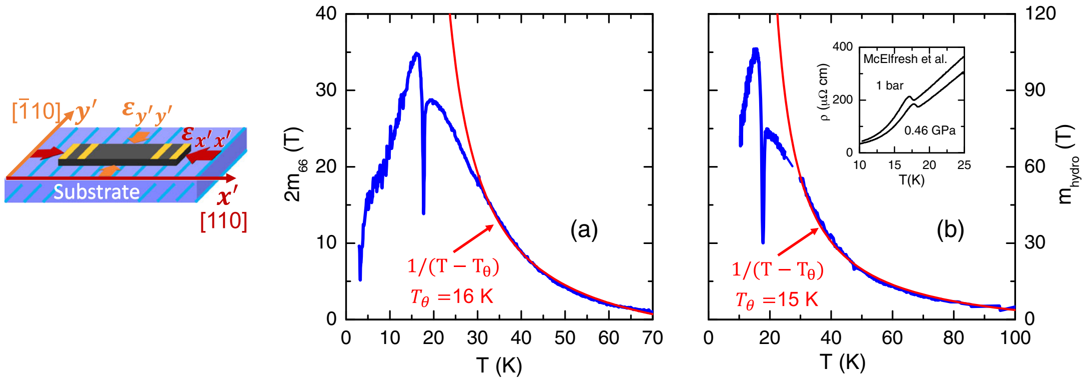

FIG. 4. Temperature dependence of the (a) uniaxial $2m_{66}$ and (b) hydrostatic $m_{hydro}$ elastoresistivity coefficients. The data in (a) were obtained using a uniaxial strain introduced by a glass-fiber substrate (see sketch on left-hand side and Ref. [22]), and $m_{hydro}$ was calculated using the difference in resistivity between pressures of 1 bar and 0.46 GPa from McElfresh *et al.* [31] shown in the inset of (b).

thermal-expansion and Young's-modulus data, is most likely not due to a nematic response.

Previous work on $URu_2Si_2$ by McElfresh *et al.* [31] clearly demonstrates that the resistivity is also highly sensitive to hydrostatic pressure. One can also define a dimensionless hydrostatic elastoresistivity coefficient as $m_{\mathrm{hydro}} = (\Delta\rho/\rho)/(\Delta V/V)$, where $\Delta\rho/\rho$ is the relative change of resistivity induced by a relative change of volume $\Delta V/V$. To calculate $m_{\mathrm{hydro}}$, we took the difference in resistivity between 1 bar and 0.46 GPa from Ref. [31] [see inset of Fig. 4(b)], and the volume change was computed using the bulk-modulus value of 190 GPa from Ref. [32]. The inferred temperature dependence of $m_{\mathrm{hydro}}(T)$, plotted in Fig. 4(b), remarkably resembles that of $2m_{66}(T)$. It can also be fit by a Curie-Weiss divergence over a similar temperature interval, and it has a similar dip at $T_{\mathrm{HO}}$. We find that the maximum value of $m_{\mathrm{hydro}}(T)$ is roughly 3 times larger than that of $2m_{66}$. Since there is no symmetry-breaking strain in a hydrostatic-pressure experiment, these data clearly demonstrate that a Curie-Weiss-like temperature dependence of an elastoresistivity component can be obtained in the absence of nematicity, and that a Curie-Weiss-like response under a symmetry-breaking strain [as in Fig. 4(a)] does not necessarily imply that the system is nematic. The obvious question now is what is the physics behind the Curie-Weiss-like temperature dependence? As clearly shown in Ref. [31], the primary effect of pressure is to move the maximum in the resistivity $T_{\mathrm{max}}$ to higher temperatures. As a consequence, the shift of $T_{\mathrm{max}}$, together with the normalization by $\rho(T)$, accidentally leads to a Curie-Weiss-like behavior of $m_{\mathrm{hydro}}(T)$. The fact that $m_{\mathrm{hydro}}(T) \approx 3 \times 2m_{66}(T)$ strongly suggests that $2m_{66}$ is probing a fraction of the hydrostatic part. A detailed analysis actually has shown that $2m_{66}$ and the isotropic in-plane strain effects are nearly equal [23,33].

In summary, we find no evidence for an electronic nematic transition associated with the HO transition in $URu_2Si_2$ using three sensitive experimental techniques. We thus conclude that $URu_2Si_2$ does not undergo a crystallographic symmetry reduction at $T_{\mathrm{HO}}$, and that the HO must be restricted to tetragonal symmetry, as proposed by the recent NMR results [9]. The HO state can thus be narrowed down to a non-nematic rank-5 multipolar order (dotriacontapolar) if time reversal symmetry breaking (TRSB) is preserved, but other possibilities exist if TRSB is broken [6,9,34,35]. Our results, which are consistent with several decades of intense investigations of this material [2], are thus in contradiction to the recent studies reporting a symmetry reduction at the HO transition. The proponents of the nematic transition may argue that our crystals are not clean enough, and that only the cleanest crystals undergo this transition. A recent paper [36] found evidence for a pressure induced symmetry breaking transition near 100 K at roughly the pressure where magnetic order sets in. The purported symmetry breaking at $T_{\mathrm{HO}}$ found in Ref. [4] was included in their phase diagram and was argued to result from a reduced *a*-axis lattice parameter in these crystals [36]. The *a*-axis lattice parameter of our typical crystals [$a = 4.1327(3)$ Å] [18], on the other hand, is larger and is thus in the stable HO region of this phase diagram. This is advantageous, since we are most interested in investigating the stable HO phase, and our results clearly demonstrate that the HO phase transition in this stable region does not induce a lattice symmetry reduction. Finally, the quantitative difference between [100] and [110] directions in elastoresistivity is intriguing, but may just result from the natural inequivalence of strain in [100] and [110] directions, which should be observable in any tetragonal system. Recent elastoresistivity measurements on tetragonal $RbFe_2As_2$ and $CsFe_2As_2$ have found huge elastoresistive responses with a Curie-Weiss-like temperature dependence indicative of nematicity [37], although previous high-resolution thermal-expansion [38,39] and shear-modulus measurements [39] find little evidence of a nematic transition or nematic fluctuations in these materials. Both $RbFe_2As_2$ and $CsFe_2As_2$, however, have large electronic Grüneiesen parameters at low temperature [38–40], and thus a highly strain sensitive electronic structure. More work using both thermodynamic as well as elastoresistive measurements on well-characterized crystals are needed to study this apparent discrepancy, in particular in cuprates, where the elastoresistive response is quite weak [41].

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[*] These authors contributed equally to this work.

[†] christoph.meingast@kit.edu

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