# **Absence of a bulk thermodynamic phase transition to a density wave phase in UTe2**

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(Received 9 July 2024; revised 11 September 2024; accepted 16 September 2024; published 9 October 2024)

Competing and intertwined orders are ubiquitous in strongly correlated electron systems, such as the charge, spin, and superconducting orders in the high-*T<sub>c</sub>* cuprates. Recent scanning tunneling microscopy (STM) measurements provide evidence for a charge density wave (CDW) that coexists with superconductivity in the heavy fermion metal UTe<sub>2</sub>. This CDW persists up to at least 7.5 K, and, as a CDW breaks the translational symmetry of the lattice, its disappearance is necessarily accompanied by a thermodynamic phase transition. We report high-precision thermodynamic measurements of the elastic moduli of UTe<sub>2</sub>. We observe no signature of a phase transition in the elastic moduli down to a level of 1 part in 107, strongly implying the absence of bulk CDW order in UTe2. We suggest that the CDW and associated pair density wave observed by STM may be confined to the surface of UTe<sub>2</sub>.

DOI: [10.1103/PhysRevB.110.144507](https://doi.org/10.1103/PhysRevB.110.144507)

# **I. INTRODUCTION**

The superconductivity of  $UT_{e_2}$  is unconventional in many respects: it is spin triplet [\[1\]](#page-4-0), it has a reentrant phase at very high magnetic fields  $[2,3]$ , and its superconducting  $T_c$ bifurcates into two transitions under pressure [\[4,5\]](#page-4-0). Recent scanning tunneling microscopy (STM) experiments [\[6–9\]](#page-4-0) have provided evidence for even more unusual behavior: the coexistence of superconductivity with an incommensurate charge density wave (CDW). Upon applying a magnetic field, superconductivity and the CDW are suppressed at the same critical field [\[6\]](#page-4-0). This observation suggests a close connection between superconductivity and the CDW in  $UT_{2}$ , potentially connected via a parent pair density wave (PDW) [\[8\]](#page-4-0).

STM measurements clearly indicate that the CDW persists up to at least 7.5 K—more than a factor of 3 higher than  $T_c$ —and disappears at a temperature no higher than 12 K [\[9\]](#page-4-0). A corollary of this observation is that, upon cooling below 12 K, UTe<sub>2</sub> first enters a broken-symmetry CDW phase, followed by the onset of superconductivity at lower temperature. As broken-symmetry phases are necessarily accompanied by thermodynamic phase transitions [\[10\]](#page-4-0), it is natural to ask whether such a phase transition is observed between 7.5 and 12 K in UTe<sub>2</sub>. The answer thus far is negative: the superconducting transition is the only phase transition visible in specific-heat measurements at ambient pressures and in zero magnetic field [\[11\]](#page-4-0).

The apparent absence of a second phase transition leads to the following question: does CDW order exist in the bulk of  $UTe<sub>2</sub>$ , or is it confined to the surface? This gets to the heart of a broader problem: which of the many exotic phenomena that have been discovered in  $UTe<sub>2</sub>$  are representative of the bulk, and which are particular to the surface? For example, the polar Kerr effect  $[12]$ , STM  $[13]$ , and microwave conductivity  $[14]$ measurements all suggest a two-component, time-reversal symmetry-breaking order parameter, whereas ultrasound [\[15\]](#page-5-0) and some specific-heat [\[16\]](#page-5-0) measurements suggest a singlecomponent order parameter. One possible resolution is that Kerr, STM, and microwaves are all sensitive to a unique superconducting state on the surface of UTe<sub>2</sub>, whereas ultrasound and specific heat are sensitive to the bulk order parameter. This issue of bulk versus surface superconductivity in  $UT_{22}$ is closely related to the existence of a CDW, as a superconducting PDW is necessarily accompanied by a CDW. The existence of such a PDW/CDW pair in the bulk would strongly constrain the microscopic mechanism of Cooper pairing in  $UTe<sub>2</sub>$ . To investigate the possibility of a phase transition to bulk CDW order in  $UTe<sub>2</sub>$ , we measure the elastic moduli as a function of temperature, from 2 to 280 K. Elastic moduli

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FIG. 1. Elastic moduli of UTe<sub>2</sub> from 2 to 280 K. The three compressional moduli (left) and shear moduli (right) are shown from 1.2 to 300 K.  $\Delta c/c$  is defined as  $[c(T) - c(2K)]/c(2K)$ . The *a*-axis resistivity of UTe<sub>2</sub> is plotted as a dashed line for comparison and is taken from Eo *et al.* [\[17\]](#page-5-0). The downturn of the resistivity around 50 K coincides with deviations from conventional stiffening due to lattice anharmonicity in  $c_{11}$ ,  $c_{22}$ ,  $c_{33}$ ,  $c_{55}$ , and  $c_{66}$ . Note that the noise in the data is smaller than the width of the lines on this scale.

are particularly sensitive to CDW phase transitions because they break the translational symmetry of the lattice. This has been investigated extensively in other CDW systems, such as the rare-earth tritellurides, where discontinuities in the elastic moduli at  $T_{CDW}$  are of order a few times  $10^{-2}$  of the total elastic moduli [\[18\]](#page-5-0). Because elastic moduli can be measured with better than one part in  $10<sup>7</sup>$  precision (see the Methods section), our measurements have many decades of sensitivity to explore the possibility of a bulk phase transition to CDW order in UTe<sub>2</sub>.

# **II. RESULTS**

Elastic moduli,  $c_{ij}$ , are the thermodynamic coefficients characterizing the susceptibility of a material to strain. In terms of the total free energy  $F$ , elastic moduli are given by

$$
c_{ij} = \frac{\partial^2 \mathcal{F}}{\partial \epsilon_{ij}^2},\tag{1}
$$

where  $\epsilon_{ij}$  is a particular component of the strain tensor. These moduli are related to the sound velocities,  $v_{ij}$ , by  $v_{ij} = \sqrt{c_{ij}/\rho}$ , where  $\rho$  is the material density. Like other thermodynamic susceptibilities, such as specific heat, elastic moduli exhibit singular behavior at phase transitions [\[19\]](#page-5-0).

We first show the elastic moduli corresponding to all six unique strains in  $UTe<sub>2</sub>$  measured over a broad temperature range—from 2 to 280 K—using pulse echo ultrasound. These data are reproduced from Theuss *et al.* [\[15\]](#page-5-0), where details about the sample preparation and experimental technique are given.

All six elastic moduli exhibit only smooth behavior across the entire temperature range down to  $T_c$ . The shear modulus *c*<sup>44</sup> exhibits conventional stiffening due to lattice anhar-monicity [\[20\]](#page-5-0). The other two shear  $(c_{55}$  and  $c_{66}$ ) and three compressional moduli  $(c_{11}, c_{22},$  and  $c_{33})$  exhibit smooth evolution with temperature that is associated with the onset of Kondo coherence near 50 K  $[21,22]$ . This demonstrates the sensitivity of five out of six elastic moduli to changes in the electronic structure of UTe<sub>2</sub>. Despite this sensitivity, no

sharp changes in slope or discontinuities that would be indicative of an electronic phase transition are visible on this scale.

To further constrain the presence or absence of a CDW transition, we performed high-resolution resonant ultrasound spectroscopy (RUS) measurements on UTe<sub>2</sub> across the temperature range where the CDW peaks disappear in the STM experiments [\[9\]](#page-4-0). RUS measures the mechanical resonance frequencies of a sample. Each resonance frequency is determined by the sample geometry, the material density, and the elastic moduli. While procedures exist for decomposing the temperature dependence of the resonance frequencies into the temperature dependence of the elastic moduli [\[23–25\]](#page-5-0) (see Methods), the highest signal to noise is obtained by directly examining the resonance frequencies.

Figure [2](#page-2-0) shows the temperature dependence of five resonance frequencies from 4 to 20 K—across the temperature range where the CDW disappears in the STM experiments. No anomalies are visible across the entire temperature range. These resonance frequencies contain admixtures of all nine elastic moduli in different proportions: the topmost resonance (blue) is dominated by compressional moduli, whereas the resonance at the bottom of the figure (green) is dominated by shear moduli. Thus, all resonances should show a singular jump, as well as a change in slope, at a phase transition [\[26\]](#page-5-0). Using the signal-to-noise of our measurement, we can constrain any singularity in the elastic moduli to be smaller than  $1 \times 10^{-7}$  (see Methods).

To provide a sense of scale for what is expected at a CDW transition, we perform similar RUS measurements on  $CsV<sub>3</sub>Sb<sub>5</sub>$ .  $CsV<sub>3</sub>Sb<sub>5</sub>$  has both a CDW transition near 90 K and a superconducting  $T_c$  near 2 K  $[27]$ . The analogy with UTe<sub>2</sub> is close, as there is also STM evidence for a PDW in this material [\[28\]](#page-5-0). The impact of the CDW phase transition on the elastic moduli is striking: discontinuities on the order of  $2 \times 10^{-2}$ are visible at *T*<sub>CDW</sub>—five orders of magnitude larger than our signal to noise in UTe<sub>2</sub>. Similar-sized anomalies at  $T_{CDW}$  are present in the elastic moduli of the rare-earth tritellurides, and anomalies of order  $10^{-4}$  are found at the transition to the

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FIG. 2. Low-temperature elastic resonances of CsV<sub>3</sub>Sb<sub>5</sub> and UTe<sub>2</sub>. High-resolution mechanical resonances of UTe<sub>2</sub> (left) and CsV<sub>3</sub>Sb<sub>5</sub> (right). Each curve tracks the frequency of a single mechanical resonance of a single-crystal sample.  $\Delta f/f$  is defined as  $[f(T) - f(T_0)]/f(T_0)$ , where  $T_0$  is the highest temperature shown. A gray bar indicates the temperature range over which the CDW disappears in STM measurements of UTe2. Note that the noise in the data is smaller than the width of the lines on this scale.

high-field CDW phase of the high- $T_c$  cuprates. These results are summarized, along with other examples from the literature, in Table I.

#### **III. DISCUSSION**

Our data constrain any thermodynamic signature of a CDW phase transition in UTe<sub>2</sub> to be  $\Delta c/c < 1 \times 10^{-7}$ . This is five orders of magnitude smaller than what is observed in  $CsV<sub>3</sub>Sb<sub>5</sub>$ and in the rare-earth tritellurides [\[18\]](#page-5-0), and three orders of magnitude smaller than what is observed in the high- $T_c$  cuprates. This constraint is two orders of magnitude tighter than that placed by previous thermal expansion [\[11](#page-4-0)[,29\]](#page-5-0) and specificheat  $[30]$  measurements on UTe<sub>2</sub> (these investigations did not discuss the absence of a CDW transition, but they do present data taken over the relevant temperature range).

There are two common arguments as to why a CDW might not exhibit a thermodynamic signature in the elastic moduli: from broadening of the transition due to disorder, and from insensitivity of the CDW to the lattice (a "purely electronic" CDW). We address these possibilities in turn.

Disorder tends to broaden all thermodynamic singularities at a CDW transition [\[31,32\]](#page-5-0). This is most apparent in the high- $T_c$  cuprates. There, CDW correlation lengths range from a few unit cells in  $Bi_2Sr_2CaCu_2O_{8+\delta}$  [\[34\]](#page-5-0) to roughly 100 Å in YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+δ</sub> [\[35–37\]](#page-5-0). Because of the short correlation length, no anomaly is found in either the specific heat or the elastic moduli at the onset of the CDW correlations [\[38,39\]](#page-5-0). Only upon applying a magnetic field is the correlation length increased to roughly 300 Å and an associated singularity observed in the elastic moduli [\[38,40\]](#page-5-0). The CDW seen by  $STM$  in UTe<sub>2</sub> is qualitatively different from that found in the cuprates: in  $\text{UTe}_2$ , the CDW peaks are of a similar width to the crystalline Bragg peaks [\[6\]](#page-4-0). Furthermore, existing ultrasound investigations into the superconducting state of  $UT_{e_2}$  reveal sharp superconducting transitions—with widths only 5% of  $T_c$ —that show no signs of broadening due to disorder [\[15\]](#page-5-0). Thus disorder is an unlikely explanation for the lack of thermodynamic singularity at a putative  $T_{CDW}$ .

A "purely electronic" CDW would be a CDW that is entirely decoupled from the crystalline lattice. To our knowledge, such a state does not exist. The very fact that  $UTe<sub>2</sub>$  has a non-Galilean-invariant (i.e., nonspherical) Fermi surface [\[41\]](#page-5-0) means that the electronic degrees of freedom are coupled to the lattice potential. As sound waves deform that lattice potential, they are necessarily coupled to the conduction electrons.

TABLE I. Elastic moduli discontinuities in several CDW materials. *Y*<sub>[ijk]</sub> is a Young's modulus measured with the compressive stress in the [ijk] direction. NbSe<sub>2</sub>, CsV<sub>3</sub>Sb<sub>5</sub>, YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6.55</sub>, and Lu<sub>5</sub>Ir<sub>4</sub>Si<sub>10</sub> also have superconducting transitions at lower temperatures. The elastic anomaly in TbTe<sub>3</sub> is representative of all rare-earth tritellurides  $[33]$ .

Material	$T_{CDW}$ (K)	$\Delta c/c$	$c_{ij}$	Reference	
UTe <sub>2</sub>	$10 - 12$	$< 1 \times 10^{-7}$		This work	
$CsV_3Sb_5$	93	$\approx$ 10 <sup>-2</sup>		This work	
TbTe <sub>3</sub>	333	$2 \times 10^{-2}$	$c_{11}$	Saint-Paul et al. [18]	
		$2 \times 10^{-2}$	$c_{33}$		
$K_{0,3}MoO3$	180	$2 \times 10^{-2}$	$Y_{[102]}$	Brill <i>et al.</i> $[42]$	
$Lu_5Ir_4Si_{10}$	80	$1 \times 10^{-2}$	$c_{11}$	Saint-Paul et al. [43]	
		$6 \times 10^{-3}$	$c_{33}$		
TTF-TCNO	53	$1 \times 10^{-2}$	$Y_{[010]}$	Barmatz et al. [44]	
$2H$ -NbSe <sub>2</sub>	30	$1 \times 10^{-3}$	$Y_{[100]}$	Barmatz et al. [45]	
$YBa_2Cu_3O_{6.55}$ ( <i>H</i> = 30 T)	50	$8 \times 10^{-5}$	$c_{22}$	Laliberté et al. [38]	

$f$ (MHz)	$\alpha_{11}$	$\alpha_{22}$	$\alpha_{33}$	$\alpha_{12}$	$\alpha_{13}$	$\alpha_{23}$	$\alpha_{44}$	$\alpha_{55}$	$\alpha_{66}$
2.44	0.141	0.088	0.212	0.004	$-0.092$	$-0.028$	0.263	0.129	0.283
3.01	0.250	0.178	0.310	$-0.022$	$-0.152$	$-0.045$	0.190	0.168	0.123
3.08	0.241	0.113	0.190	$-0.030$	$-0.115$	$-0.007$	0.233	0.129	0.246
3.20	0.191	0.082	0.244	$-0.004$	$-0.134$	$-0.020$	0.246	0.146	0.249
3.45	0.212	0.108	0.190	$-0.015$	$-0.109$	$-0.018$	0.248	0.110	0.274

<span id="page-3-0"></span>TABLE II. Resonance frequency composition. The coefficient of each elastic modulus that makes up the resonance frequency as defined in Eq. (2).

This is evidenced by softening of the elastic moduli at the onset of Kondo coherence near 50 K (Fig. [1\)](#page-1-0), as well as by the sharp thermodynamic singularities seen at  $T_c$  [\[15\]](#page-5-0). Any redistribution of the charge density in a metal is necessarily compensated for by a displacement of the lattice to maintain local charge neutrality, and thus a thermodynamic signature in the elastic moduli at a CDW transition is inescapable.

We find no evidence for a transition to a CDW phase in the bulk of  $UTe<sub>2</sub>$ , despite such evidence existing on the [011] surface as measured by STM  $[6-9]$ . Indeed, there are other examples of CDWs that exist only on the surfaces of metals [\[46–48\]](#page-6-0). Our observation is also consistent with the lack of a CDW phase transition occurring in other bulk-sensitive probes such as heat capacity [\[30\]](#page-5-0), thermal expansion  $[11,29]$  $[11,29]$ , and NMR  $[49]$ . We also note that recent x-ray diffraction measurements have failed to detect the requisite superlattice peaks expected for a CDW [\[50\]](#page-6-0). This suggests that the surface of  $UTe<sub>2</sub>$  may host different ordered states from the bulk, including possibly a different superconducting order parameter. Thus, while the host of unconventional phenomena discovered by both bulk and surface measurements in  $UTe<sub>2</sub>$  must ultimately be understood within a single framework, one should be careful when extrapolating observations made on the surface to the bulk and vice versa.

### **IV. METHODS**

#### **A. Sample preparation**

#### 1.  $CsV_3Sb_5$

Single crystals of  $CsV_3Sb_5$  were synthesized by the selfflux method in an inert environment. Elemental liquid Cs (Alfa 99.98%), V powder (Sigma 99.9%) in-house prepurified in a 9:1 ethanol/hydrochloric acid mixture, and Sb shot (Alfa 99.999%) were weighed out to  $Cs_{20}V_{15}Sb_{120}$  stoichiometry and milled in a tungsten carbide vial. The precursor milled powder was heated up to 1000 ◦C, soaked for 12 h, cooled down to 900 ◦C at 5 ◦C/h, and further cooled down to 500 ◦C at  $2^{\circ}$ C. Once at room temperature, the resulting crystals were extracted manually in air.

A single-crystal specimen was selected for RUS based on visual inspection. Samples with cracks, excessive flux, and intergrowth of secondary phases were avoided due to their detrimental effects on the mechanical quality factors. The sample chosen for the RUS measurements presented here is irregularly shaped, with two parallel faces perpendicular to the crystallographic *c* axis. Its dimensions are roughly

 $0.5 \times 0.8$  mm<sup>2</sup> in the *a-b* plane and 0.09 mm along the *c* axis.

# 2. **UTe**<sub>2</sub>

Single crystals of  $UTe<sub>2</sub>$  were grown by the chemical vapor transport method as described in Ran *et al.* [\[51\]](#page-6-0).

The sample used for the RUS measurement was a roughly  $1 \times 1 \times 1$  mm<sup>3</sup> large, irregularly shaped sample. More details on the shape of this sample are found in Theuss *et al.* [\[24\]](#page-5-0) under Sample B.

The surface of a single-crystal specimen was digitized using a Zeiss Xradia Versa XRM-520 X-ray nano-CT, and the digitized mesh was aligned to the crystal axes using backreflection Laue.

#### **B. Experiments**

### *1. Pulse-echo ultrasound*

Measurements were performed in an Oxford Instruments Heliox 3He refrigerator using a traditional phase-comparison pulse-echo method. Measurements were performed on three different samples in six different transducer configurations. More details about the samples and experiment can be found in the Methods section of Theuss *et al.* [\[15\]](#page-5-0).

### *2. Resonant ultrasound spectroscopy*

We performed RUS on a single-crystal sample of  $UTe<sub>2</sub>$ in a custom-built probe immersed in a bath of  ${}^{4}$ He. Details of the apparatus and the procedure for measuring resonance frequencies can be found in the Methods section of Ghosh *et al.* [\[52\]](#page-6-0). Temperature sweeps were performed using a slow ramp rate of approximately 0.025 K/min.

We fit the elastic moduli of this sample at 4 K using the method described in Theuss *et al.* [\[24\]](#page-5-0). The absolute elastic moduli for this sample are found in Table III of Theuss *et al.* [\[24\]](#page-5-0) under Sample B. The raw resonance frequencies, as well as the fit we obtain, are found in Table XI of the supplemental material to Theuss *et al.* [\[24\]](#page-5-0).

The fit to the resonance spectrum allows us to determine the contribution from each elastic modulus to each resonance frequency. Each resonance frequency  $f_k$  contains contributions from all elastic moduli, and the relative change in resonance frequency as a function of temperature is given by

$$
\frac{\Delta f_k}{f_k} = \sum_{i,j} \alpha_{ij}^{(k)} \frac{\Delta c_{ij}}{c_{ij}},\tag{2}
$$

<span id="page-4-0"></span>

FIG. 3. Noise level of  $UTe<sub>2</sub>$  resonances. The same data as presented in Fig. [2](#page-2-0) but with a fifth-order polynomial background subtracted. The spacing between points is approximately 8 mK, and the data have had a moving average applied over a 10-point window. The data have been offset vertically for clarity.

where the  $\alpha_{ij}^{(k)}$  coefficients are temperature-independent,  $\Delta f_k / f_k = [f_k(T) - f_k(T_0)] / f_k(T_0)$  is the relative change in resonance frequency referenced to temperature  $T_0$ , and likewise for  $\Delta c_{ij}/c_{ij}$ . The coefficients  $\alpha_{ij}^{(k)}$  sum to 1 for each resonance:  $\sum_{i,j} \alpha_{ij}^{(k)} = 1$ .

Figure [2](#page-2-0) shows five resonance frequencies selected for having high *Q* factors  $(>10^5)$  and qualitatively different temperature dependencies such that all elastic moduli are represented in this data set. This representation is quantified through the  $\alpha_{ij}^{(k)}$  coefficients of Eq. [\(2\)](#page-3-0). Table [II](#page-3-0) shows these coefficients for the five resonance frequencies plotted in Fig. [2.](#page-2-0)

Discontinuities are expected in the three compressional elastic moduli— $c_{11}$ ,  $c_{22}$ , and  $c_{33}$ —at any phase transition [\[52\]](#page-6-0),

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irrespective of which symmetries are broken (shear moduli can have discontinuities for some, but not all order parameters). Thus, it is critical that the three compressional moduli are well-represented in the resonance frequencies we analyze. Table [II](#page-3-0) shows that the temperature dependences of  $c_{11}$ ,  $c_{22}$ , and *c*<sup>33</sup> each make up between 10% and 30% of the total frequency shift for the five resonances shown in Fig. [2.](#page-2-0) Thus if there was a phase transition, these five frequencies would show it.

### **C. Noise analysis**

Figure  $3$  shows the same UTe<sub>2</sub> RUS data from Fig. [2,](#page-2-0) but with a fifth-order polynomial subtracted and a moving average of 10 points applied. With a temperature step of approximately 8 mK, this averages over an 80 mK window—the same width as the thermodynamic singularity at  $T_c$  in UTe<sub>2</sub> [\[15\]](#page-5-0). The noise on this scale is of order  $\Delta f / f \approx \pm 1 \times 10^{-7}$ .

# **ACKNOWLEDGMENTS**

A.S., B.J.R., and F.T. acknowledge funding from the U.S. Department of Energy Office of Basic Energy Sciences under Award No. DE-SC0020143 (ultrasound experiments and analysis). N.B. and J.P. acknowledge support from the Department of Energy Award No. DE-SC-0019154 (sample characterization), the Gordon and Betty Moore Foundation's EPiQS Initiative through Grant No. GBMF9071 (materials synthesis), the National Science Foundation under Grant No. DMR-2105191 (sample preparation), and the Maryland Quantum Materials Center and the National Institute of Standards and Technology. B.J.R. and F.T. acknowledge use of the Cornell Center for Materials Research Shared Facilities, which are supported through the NSF MRSEC program (DMR-1719875).

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