Magnetoelastic coupling in URu$_2$Si$_2$: probing multipolar correlations in the hidden order state

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Time reversal symmetry and magnetoelastic correlations are probed by means of high-resolution volume dilatometry in URu$_2$Si$_2$ at cryogenic temperatures and magnetic fields more than enough to suppress the hidden order state at $H_{HO}(T = 0.66 \text{ K}) \approx 35 \text{ T}$. We report a significant crystal lattice volume expansion at and above $H_{HO}(T)$, and even above $T_{HO}$, possibly a consequence of field-induced $f$-electron localization, and hysteresis at some high field phase boundaries that confirm volume involvement. We investigate in detail the magnetoestriction and magnetization as the temperature is reduced over two decades from 50 K where the system is paramagnetic, to 0.5 K in the realms of the hidden order state. We find a dominant quadratic-in-field dependence $\Delta L/L \propto H^2$, a result consistent with a state that is symmetric under time reversal. The data shows, however, an incipient yet unmistakable asymptotic approach to linear $(\Delta L/L \propto 1 - H/H_0)$ for 15 T $< H < H_{HO}(0.66 \text{ K}) \approx 35 \text{ T}$ at the lowest temperatures. We discuss these results in the framework of a Ginzburg-Landau formalism that proposes a complex order parameter for the HO to model the (H,T,p) phase diagram.

Despite decades of research, URu$_2$Si$_2$ remains among the most fascinating and puzzling of correlated electron systems [1]. At the focus of the puzzle is the appearance of an ordered phase, heralded by a large specific heat anomaly at 17 K. The nature of the order underlying this phase remains ambiguous, hence the term hidden order (HO) phase. There have been numerous theoretical attempts to close the loop and many experimental probes to distinguish among them. These approaches can be divided into two classes: one which assumes that the material is primarily a band metal, with the $U$ electrons fully hybridized with band electrons derived from Ru and Si orbitals, the other assuming that the U-atom configuration is $5f^2$ with the HO phase evolving from either singlet or doublet crystal field ground states. There is ample experimental evidence to support each approach. Both band and CEF approaches can explain the strong singlet or doublet crystal field ground states. There is some evidence in the literature for breaking of time reversal symmetry (NMR [3], [4], SR [5] and Kerr rotation [6]). One objective of this work is to carry the study of magnetoestriction to high magnetic fields in a search for broken time-reversal symmetry. We are further motivated by the recent observation by Kung et al. [7] of a sharp feature in Raman scattering with $A_{2g}$ symmetry which appears below the HO transition $T_{HO} = 17.5 \text{ K}$. That feature has been tracked by Buhot [9] as a function of applied field and found to decrease in strength toward the HO critical field $H_{HO} = 34 \text{ T}$. The Raman feature was demonstrated to be consistent with the electric-hexadecapole order parameter proposed by Haule and Kotliar [10] and by Kusunose and Harima [11]. The detailed interaction between the proposed hexadecapole order and thermal expansion was calculated in mean-field theory by Haule and Kotliar. We extend that model to treat magnetostriction and demonstrate that it explains, in detail, the asymptotic approach toward linear-in-field magnetostriction reported here. We have also measured the magnetization at various low temperatures to 34 T and find it to be strikingly linear in field with no apparent correlation to the magnetostriction. Our pulsed field measurements reproduce the rich cascade of low-temperature, high-field phases [7-11] seen above $H_{HO}$. None of the results reported here, including the asymptotic approach to linear-in-field magnetostriction, supports broken time-reversal symmetry in the HO phase. Indeed, the proposed hexadecapole ordering preserves that symmetry.

We use an optical fiber Bragg grating-based dilatometry technique described before [12]. Single crystal samples of URu$_2$Si$_2$ were grown by the Czochralski technique, described elsewhere [6], and oriented by Laue diffraction in backscattering geometry. Bar-shaped samples were cut of approximate dimensions $2\times0.5\times0.5 \text{ mm}^3$ with the longest dimension along the principal crystallographic axis $a$ or $c$. Axial strain is obtained when the fiber is mounted parallel to the applied field, transverse strain...
can be measured when the sample space in the magnet permits bending the Bragg-grating-furbished end of the fiber perpendicular to the magnetic field without losing the internal reflection condition. A resolution $\Delta L/L \sim 1$ part per million (ppm) is achieved in pulsed fields and $\sim 0.03$ ppm in continuous fields [12–14]. Complementary magnetization measurements were accomplished using a sample extraction method. The ultimate resolution in this technique benefits from pulsed magnetic fields. Measurements were carried out in 60 T and 65 T pulsed (2.5 sec and 35 ms long respectively) and in 35 T continuous electromagnets, at the NHMFL.

Fig. 1 shows a combination of field (main panels) and temperature (insets) dependent dilatometry data. The insets in panels (a-c) show zero field (H=0) strain $\Delta L/L$ vs. T where $\Delta L/L(T) = (L(T)-L(25K))/L(25K)$ in units of ppm. In panel (a) inset, we see that the $a$-axis [010] shrinks with a decrease in temperature, displaying an anomaly upon entering the HO phase (see arrow). The $c$-axis [001] in panel (b) inset, however, expands as expected from the Poisson rule. The volume effect is calculated as $\Delta V/V = \Delta c/c + 2\Delta a/a$ and displayed in panel (c) inset. The computation is justified by the fact that the material is tetragonal. The coefficient of volumetric thermal expansion $\beta(T) = \partial(\Delta V/V)/\partial T = \alpha_c(T) + 2\alpha_a(T)$ (not shown) is in close agreement with earlier data [25]. Main panels show the magnetostriction $\Delta L/L$ vs. magnetic field $H \parallel$ [001] at various temperatures with the $a$-axis, panel (a), expanding and the $c$-axis, panel (b), contracting in fields large enough to suppress the HO. An important lattice effect is observed even at $T = 20$ K, above $T_{\text{HO}}=17$ K. We calculate the magneto-volume change in field in panel (c). At base temperature $T = 1.3$ K we see very small changes at low fields, until reaching $H_{\text{HO}} \approx 35$ T when the HO is suppressed. At this field we see a large increase in volume, significantly bigger than observed upon cooling at $T_{\text{HO}}$ in the thermal expansion data, indicating the transition to phases II and III, as defined in the literature [16–19]. We finally see a break in slope and a continued increase in volume at high fields, defined in the literature [16–19]. We calculate the magneto-volume change in field in panel (c). At base temperature $T = 1.3$ K we see very small changes at low fields, until reaching $H_{\text{HO}} \approx 35$ T when the HO is suppressed. At this field we see a large increase in volume, significantly bigger than observed upon cooling at $T_{\text{HO}}$ in the thermal expansion data, indicating the transition to phases II and III, as defined in the literature [16–19]. We finally see a break in slope and a continued increase in volume at high fields, defined in the literature [16–19]. We calculate the magneto-volume change in field in panel (c). At base temperature $T = 1.3$ K we see very small changes at low fields, until reaching $H_{\text{HO}} \approx 35$ T when the HO is suppressed. At this field we see a large increase in volume, significantly bigger than observed upon cooling at $T_{\text{HO}}$ in the thermal expansion data, indicating the transition to phases II and III, as defined in the literature [16–19]. We finally see a break in slope and a continued increase in volume at high fields, defined in the literature [16–19]. We calculate the magneto-volume change in field in panel (c). At base temperature $T = 1.3$ K we see very small changes at low fields, until reaching $H_{\text{HO}} \approx 35$ T when the HO is suppressed. At this field we see a large increase in volume, significantly bigger than observed upon cooling at $T_{\text{HO}}$ in the thermal expansion data, indicating the transition to phases II and III, as defined in the literature [16–19]. We finally see a break in slope and a continued increase in volume at high fields, defined in the literature [16–19]. We calculate the magneto-volume change in field in panel (c). At base temperature $T = 1.3$ K we see very small changes at low fields, until reaching $H_{\text{HO}} \approx 35$ T when the HO is suppressed. At this field we see a large increase in volume, significantly bigger than observed upon cooling at $T_{\text{HO}}$ in the thermal expansion data, indicating the transition to phases II and III, as defined in the literature [16–19]. We finally see a break in slope and a continued increase in volume at high fields, defined in the literature [16–19]. We calculate the magneto-volume change in field in panel (c). At base temperature $T = 1.3$ K we see very small changes at low fields, until reaching $H_{\text{HO}} \approx 35$ T when the HO is suppressed. At this field we see a large increase in volume, significantly bigger than observed upon cooling at $T_{\text{HO}}$ in the thermal expansion data, indicating the transition to phases II and III, as defined in the literature [16–19]. We finally see a break in slope and a continued increase in volume at high fields, defined in the literature [16–19].

The field at which the volume magnetostriction curves taken at different temperatures cross, running into each other, marks the point where the coefficient of volumetric thermal expansion $\alpha_V$ changes sign [20]. Because $\alpha_V \propto V^{-1}(\partial S/\partial B)_T$ the sign change indicates accumulation of entropy that precedes a quantum critical endpoint (QCEP) [22]. These results confirm linear expansion data by Correa et al. [21]. This putative QCEP was never found in URu$_2$Si$_2$ and is presumed avoided by the presence of so-called phase III [17, 18], resembling the case of Sr$_3$Ru$_2$O$_7$ [23]. The large (>150 ppm) magneto-volume expansion observed overall, whether URu$_2$Si$_2$ is in the PM or HO state in high magnetic fields, is highly suggestive of $f$-electron localization-driven effect. Indeed, in the Kondo or partially-arrested Kondo state that develops out of a doubly degenerate ground state at low temperatures [24], the increasing magnetic field makes the transfer of atomic $f$-electron weight to conduction electrons less favorable. Direct exchange interaction among them, hence, likely results in a swollen unit cell volume. The more abrupt changes observed as the HO phase is suppressed point to a very strong anticorrelation with the degree of localization, i.e. the HO benefits from a certain degree of itinerancy and protects it but perishes at high fields, where localization is favored. As discussed below an alternative explanation includes the suppression of dipole moments on the U ions due to the onset of multipolar order for $T < T_{\text{HO}}$.

Fig. 2 shows the axial $\Delta c/c$ vs magnetic field up to 45 T measured at 0.66 K, 1.25 K, and 4 K. In this plot we can see a hysteretic region upon entering phase V at $\mu_0 H \approx 36-37$ T. This hysteresis, previously reported from transport and magnetization measurements [19, 26], confirms its bulk nature. Similar history dependent results have been observed at a proposed field-induced Lifshitz
transition in UPt$_3$Si$_2$ [24]. The hysteresis at lower fields (H=H$_{HO}$, T = 4K, red line) has a different origin. We observe that it depends strongly on the magnetic field sweep rate, becoming smaller to the point of vanishing for slower field rates and stronger thermal link to the bath below 2.1K. These observations indicate that it originates in the magnetocaloric effect [16], and is not intrinsic to the HO phase boundary.

Also intriguing in our data is a region in magnetic fields 15 T < $\mu_0$H < H$_{HO}$ where the magnetostriction appears to be remarkably linear in field, observed before by Correa at al. [21] but never before followed in detail to the zero field limit. Fig. 3 illustrates the evolution of the c-axis magnetostriction vs $H$ (left panel), and vs $H^2$ (right panel), with temperature in the HO phase. At high temperatures, well above $T_{HO}$, the magnetostriction follows very closely a quadratic field dependence. The low-temperature data (0.66 K, 1.3 K, 4 K) deviate from $H^2$ as the field increases. Indeed, the lowest temperature data appear to follow a simple hyperbolic function $\Delta c/c = 1 - \sqrt{H/H_0}$ which is asymptotic to 1-$|H/H_0|$ at $H \gg H_0$. This form is, of course, even under time reversal. Intermediate temperatures (7.5 K and 12.5 K) appear to transition between the two regimes. Deviation of the pulsed-field (blue) from steady-field (orange) data is a consequence of the magnetocaloric effect [16].

The low-temperature hyperbolic form can be traced back to a Landau-Ginzburg theory for URu$_2$Si$_2$ proposed by Haule and Kotliar [10] to account for the effects of applied magnetic fields and hydrostatic pressures. The theory considers the competition between antiferromagnetism and the hidden hexadecapole order phase by means of a complex order parameter of the form $\Psi_{HO} + i\Psi_{AF}$. The relevant coupling constants $J_{AF}$ and $J_{HO}$ are set by $T_{HO} = \Delta/(2 \arctanh(\Delta/J_{HO})) = 17.7$ K and by $T_{AF} = 15.7$ K for $J_{AF}$. The crystal field splitting of the low-lying singlets, determined from LDA+DMFT calculations, is set at $\Delta = 35$ K. The applied magnetic field is converted to temperature units via $b = 1.25 \mu_B \mu_0 H/k_B$. This set of parameters predicts a critical field separating the HO and paramagnetic phases at $\mu_0 H_{HO} = 36$ T, in agreement with the data in Fig. 1.

On entering the HO phase, a spontaneous c-axis strain appears, predicted to be

$$\epsilon_{zz} = -\frac{g_{HO} c_{13} J_{HO} \Psi_{HO}^2}{((c_{11} + c_{12})c_{33} + 2c_{13}^2) \beta} \left(\frac{c_{33}}{c_{11} + c_{12}}\right)^2$$

(1)

where $g_{HO}$ reflects the strain dependence of $J_{HO}$ and positive values of $\epsilon_{zz}$ correspond to compression. The order parameter in the limit where a Landau-Ginzburg expansion is valid is given by

$$\Psi_{HO}^2(b, T) = \frac{J_{HO} - 2a(b, T)}{4u(T)}$$

(2)

where $a(b, T) = (\Delta \lambda(b)/2) \coth(\beta\lambda(b)\Delta/2)$, $u(T) = (\Delta/8)(\sinh(\beta\Delta) - \beta\Delta)(\cosh^2(\beta\Delta/2))/\sinh^2(\beta\Delta/2)$ and

$$\lambda(b) = \sqrt{1 + \left(\frac{2b}{\Delta}\right)^2 \left(\frac{J_{HO}}{J_{HO} + J_{AF}}\right)^2}$$

(3)

Note that, at low temperatures, the hyperbolic cotangent in the expression for $a(b, T)$ approaches unity and the magnetostriction then follows the simple hyperbolic law, with $H_0$ identified as

$$\mu_0 H_0 = \left(\frac{k_B \Delta}{2.5 \mu_B}\right) \left(\frac{J_{HO} + J_{AF}}{J_{HO}}\right) = 40$$(4)
The hidden order state is nonmagnetic and therefore is not directly coupled to the magnetic field. The AFM order $\Psi_{AF}$, which vanishes at zero field in the HO phase, acquires a non-zero value upon the application of a field, reflected by the appearance of $J_{AF}$ in $\lambda(b)$. The above expression for the order parameter vanishes at $T_{HO}$ in agreement with the prediction of Haule and Kotliar.

To compare with the experiment, we calculate

$$\Psi_{HO}^2(b, T) - \Psi_{HO}^2(0, T) = \frac{a(0, T) - a(b, T)}{2a(T)} \mu_0 H(T)$$

and treat $g_{HO}$ as an adjustable parameter to fit the field dependence of $\mu_0 H(T)$. Combining the elastic constants reported by B. Wolf, et al. [29], with $J_{HO} = 3.9 \times 10^7$ erg/cm³ (from $T_{HO}$ and the U-atom density), we find the magnetostrictive strain to be

$$\Delta c/c = 8.4 g_{HO}(\Psi_{HO}^2(b, T) - \Psi_{HO}^2(0, T))$$

Clearly, the onset of $\Psi_{HO}^2 > 0$ first increases the c-axis lattice parameter on cooling, and then causes magnetostriction as $\Psi_{HO}^2 \rightarrow 0$ in high fields. With the calculated $\Psi_{HO}^2(0) = 0.32$ and the measured high-field $\Delta c/c = -25.4$ ppm, the coupling strength is $g_{HO} \sim 9.4$. The calculated points are shown as green asterisks in Fig. 3, following the experimental curves closely. This value is consistent with the c-axis thermal expansion between 20 K and base temperature ($\sim 20$ ppm), as seen in Fig. 1. The hidden-order exchange energy increases under hydrostatic pressure as $J_{HO}(P) = J_{HO}(1 + g_{HO}P/c_b)$, where $c_b$ is the bulk modulus. Our value predicts a consequent increase in $T_{HO}$ of 2 K at 1 GPa, which compares well with a reported value of 1.7 K/GPa [30].

One important consequence of broken time-reversal symmetry in a magnetic system is that linear magnetostriction $\Delta L/L \propto H$ is allowed [28]. Another consequence is piezomagnetism, i.e. a magnetization that is proportional to magnetostriction. We carried out magnetization measurements in pulsed magnetic fields to probe piezomagnetism. Results of these measurements are displayed in Fig. 4. The observed magnetization in the HO phase is close to linearly proportional to the magnetic field, with no evidence of $H^2$ dependence as observed in the magnetostriction [32]. This finding amounts, again, to the absence of evidence for broken time-reversal symmetry in URu$_2$Si$_2$. Materials that exhibit piezomagnetism also show strain hysteresis loops [28]. Our attempts to detect such loops yielded no indication of a spontaneous moment in zero-field- or in field-cooled conditions.

In summary, we measured the volume magnetostriction and magnetization of URu$_2$Si$_2$ to magnetic fields large enough to suppress the hidden order state. The large observed volume magnetostriction points to field-induced localization of f-electrons at high fields, with a clear sign change in the coefficient the thermal expansion that signals accumulation of entropy. The low field magnetostriction is predominantly proportional to $H^2$ at all temperatures, above and below $T_{HO}$, ruling out a spontaneous ordered magnetic moment by a time reversal symmetry argument. The magnetization M(H) shows a dominant linear dependence in the HO state, ruling out piezomagnetism. Consequently, no direct or indirect evidence for broken time-reversal symmetry is revealed. We found a low temperature c-axis magnetostriction that follows a hyperbolic function of the field, asymptotically approaching a linear dependence, in agreement with a phenomenological mean-field G-L model in the HO state that proposes a complex order parameter to model the (H,T,p) phase diagram of URu$_2$Si$_2$.

There remain questions about the exact nature of the HO phase. While Raman scattering results reveal $A_{2g}$ symmetry pointing to hexadecapolar order [7] [8], recent Ru-NQR measurements show 4-fold symmetry at the Ru and Si sites. Electric dotriacontapolar order (A$_{1u}$), with involvement of 5f and 6d electrons, was then proposed [33]. A recent ultrasonic determination of the elastic constants [2] reported that the temperature and field dependence of $c_{11} - c_{12}$ are consistent with $A_{2g}$ symmetry, but $c_{66}$ is not. XAS and RIXS [34] and NIXS [35] studies support a singlet-ground state model, as assumed by Haule and Kohtlar. We must note, however, that no pseudo-scalar order parameter, either $A_{2g}$ or $A_{1u}$-type, can by itself explain the here observed non-trivial shear response to magnetic fields. The tight connection between hexadecapolar HO order and the high-field, local moment antferromagnetic state in the form of a complex order parameter in the model is critical. We cannot rule out that a similar argument can be made for dotriacontapolar HO order, and await for the development of a complementary mean-field theory that can be contrasted against our results.
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