Elastic properties of hidden order in $\text{URu}_2\text{Si}_2$ are reproduced by modulated $B_{1g}$ order

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We develop a phenomenological mean field theory for the strain in $\text{URu}_2\text{Si}_2$ through its hidden order transition. Several experimental features are reproduced when the order parameter has $B_{1g}$ symmetry: the topology of the temperature–pressure phase diagram, the response of the strain stiffness tensor above the hidden-order transition at zero pressure, and orthorhombic symmetry breaking in the high-pressure antiferromagnetic phase. In this scenario, the hidden order is characterized by the order parameter in the high-pressure antiferromagnetic phase modulated along the symmetry axis, and the triple point joining those two phases with the paramagnetic phase is a Lifshitz point.

The study of phase transitions is central to condensed matter physics. Phase transitions are often accompanied by a change in symmetry whose emergence can be described by the condensation of an order parameter (OP) that breaks the same symmetries. Near a continuous phase transition, the physics of the OP can often be qualitatively and sometimes quantitatively described by Landau–Ginzburg mean field theories. These depend on little more than the symmetries of the OP, and coincidence of their predictions with experimental signatures of the OP is evidence of the symmetry of the corresponding ordered state.

A paradigmatic example of a material with an ordered state whose broken symmetry remains unknown is in $\text{URu}_2\text{Si}_2$. $\text{URu}_2\text{Si}_2$ is a heavy fermion superconductor in which superconductivity condenses out of a symmetry broken state referred to as hidden order (HO) [1], and at sufficiently large hydrostatic pressures, both give way to local moment antiferromagnetism (AFM). Despite over thirty years of effort, the symmetry of the HO state remains unknown, and modern theories [2–19] propose a variety of possibilities. Many of these theories rely on the formulation of a microscopic model for the HO state, but since there has not been direct experimental observation of the broken symmetry, none can be confirmed.

Recent work that studied the HO transition using resonant ultrasound spectroscopy (RUS) was able to shed light on the symmetry of the ordered state without the formulation of any microscopic model [20]. RUS is an experimental technique that measures mechanical resonances of a sample. These resonances contain information about the sample’s full strain stiffness tensor. Moreover, the frequency locations of the resonances are sensitive to symmetry breaking at an electronic phase transition due to electron-phonon coupling [21]. Ref. [20] uses this information to place strict thermodynamic bounds on the dimension of the HO OP independent of any microscopic model.

Motivated by these results, we construct a phenomenological mean field theory for an arbitrary OP coupled to strain and the determine the effect of its phase transitions on the elastic response in different symmetry channels. We find that only one OP symmetry reproduces the anomalous features of the experimental strain stiffness. That theory associates the HO state with a $B_{1g}$ OP modulated along the rotation axis, the AFM state with uniform $B_{1g}$ order, and the triple point between them with a Lifshitz point. Besides the agreement with RUS data in the HO state, the theory predicts uniform $B_{1g}$ strain in the AFM state, which was recently seen in x-ray scattering experiments [22]. The theory’s implications for the dependence of the strain stiffness on pressure and doping strongly motivates future RUS experiments that could either further support or falsify it.

The point group of $\text{URu}_2\text{Si}_2$ is $D_{4h}$, and any coarse-grained theory must locally respect this symmetry. Our phenomenological free energy density contains three parts: the free energy for the strain, the OP, and their interaction. The most general quadratic free energy of the strain $\epsilon$ is $f_{\text{ELASTIC}} = C_{ijkl} \epsilon_{ij} \epsilon_{kl}$, but the form of the bare strain stiffness tensor $C$ tensor is constrained by both the index symmetry of the strain tensor and by the point group symmetry [23]. The six independent components of strain can written as linear combinations that each behave like irreducible representations under the action of the point group, or

$$
\epsilon^{(1)}_{A_{1g}} = \epsilon_{11} + \epsilon_{22}, \quad \epsilon^{(2)}_{A_{1g}} = \epsilon_{33},
\epsilon^{(1)}_{B_{1g}} = \epsilon_{11} - \epsilon_{22}, \quad \epsilon^{(1)}_{B_{2g}} = 2\epsilon_{12},
\epsilon^{(1)}_{E_g} = 2\{\epsilon_{11}, \epsilon_{22}\}.
$$

All quadratic combinations of these irreducible strains that transform like $A_{1g}$ are included in the free energy,

$$
f_{\text{ELASTIC}} = \frac{1}{2} \sum_X C^{(ij)}_X \epsilon^{(i)}_X \epsilon^{(j)}_X,
$$

where the sum is over irreducible representations of the point group and the bare stiffnesses $C^{(ij)}_X$ are

$$
C^{(11)}_{A_{1g}} = \frac{1}{2}(C_{1111} + C_{1122}), \quad C^{(11)}_{A_{1g}} = C_{3333},
C^{(12)}_{A_{1g}} = C_{1133}, \quad C^{(1)}_{B_{1g}} = \frac{1}{2}(C_{1111} - C_{1122}),
C^{(11)}_{B_{2g}} = C_{1212}, \quad C^{(1)}_{E_g} = C_{1313}.
$$
The interaction between strain and an OP $\eta$ depends on the representation of the point group that $\eta$ transforms as. If this representation is $X$, the most general coupling to linear order is

$$f_{\text{INT}} = -bj(\epsilon)^{(i)} \epsilon_X^{(i)} \eta. \quad (4)$$

If the representation $X$ is not present in the strain there can be no linear coupling, and the effect of the OP condensing at a continuous phase transition is to produce a jump in the $A_{1g}$ strain stiffness if $\eta$ is single-component [21, 24, 25], and jumps in other strain stiffnesses if multicomponent [20]. Because we are interested in physics that anticipates the phase transition, we will focus our attention on OP symmetries that can produce linear couplings to strain. Looking at the components present in (1), this rules out all of the u-reps (which are odd under inversion) and the $A_{2g}$ irrep.

If the OP transforms like $A_{1g}$, odd terms are allowed in its free energy and any transition will be abrupt and not continuous without fine-tuning. Since this is not a feature of URu$_2$Si$_2$ Ho physics, we will henceforth rule it out as well. For $X$ as any of $B_{1g}$, $B_{2g}$, or $E_g$, the most general quadratic free energy density is

$$f_{\text{OP}} = \frac{1}{2} \epsilon^2 \eta^2 + c_{\|} (\nabla_{\|} \eta)^2 + c_{\perp} (\nabla_{\perp} \eta)^2 + D_{\perp} (\nabla_{\perp}^2 \eta)^2 + u \nabla^4 \eta \quad (5)$$

where $\nabla_{\|} = \{\partial_1, \partial_2\}$ transforms like $E_u$ and $\nabla_{\perp} = \partial_3$ transforms like $A_{2u}$. Other quartic terms are allowed—especially many for an $E_g$ OP—but we have included only those terms necessary for stability when either $r$ or $c_{\perp}$ become negative. The full free energy functional of $\eta$ and $\epsilon$ is

$$F[\eta, \epsilon] = F_{\text{OP}}[\eta] + F_{\text{ELASTIC}}[\epsilon] + F_{\text{INT}}[\eta, \epsilon] = \int dx \left( f_{\text{OP}} + f_{\text{ELASTIC}} + f_{\text{INT}} \right) \quad (6)$$

The only strain relevant to the OP is $\epsilon_X$, which can be traced out of the problem exactly in mean field theory. Extremizing with respect to $\epsilon_X$,

$$0 = \frac{\delta F[\eta, \epsilon]}{\delta \epsilon_X(x)} \bigg|_{\epsilon = \epsilon_*} = C_X \epsilon_*^2 \eta(x) - b \eta(x) \quad (7)$$

gives the optimized strain conditional on the OP as $\epsilon_*^2 \eta(x) = (b/C_X) \eta(x)$ and $\epsilon_*^2 \eta = 0$ for all other $Y$. Upon substitution into the free energy, the resulting effective free energy $F[\eta, \epsilon_*, \eta]$ has a density identical to $f_{\text{OP}}$ with $r \rightarrow \tilde{r} = r - b^2/2C_X$.

With the strain traced out, (5) describes the theory of a Lifshitz point at $\tilde{r} = c_{\perp} = 0$ [26, 27]. For a one-component OP ($B_{1g}$ or $B_{2g}$) it is traditional to make the field ansatz $\langle \eta(x) \rangle = \eta_* \cos(q_* x_3)$. For $\tilde{r} > 0$ and $c_{\perp} > 0$, or $\tilde{r} > c_{\perp}^2/4D_{\perp}$ and $c_{\perp} < 0$, the only stable solution is $\eta_* = q_* = 0$ and the system is unordered. For $\tilde{r} < 0$ there are free energy minima for $q_* = 0$ and $\eta_*^2 = -\tilde{r}/4u$ and this system has uniform order. For $c_{\perp} < 0$ and $\tilde{r} < c_{\perp}^2/4D_{\perp}$ there are free energy minima for $q_*^2 = -c_{\perp}/2D_{\perp}$ and

$$\eta_*^2 = \frac{c_{\perp}^2 - 4D_{\perp}\tilde{r}}{12D_{\perp}u} = \frac{\tilde{r}_0 - \tilde{r}}{3u} = \frac{|\Delta r|}{3u} \quad (8)$$

with $\tilde{r}_0 = c_{\perp}^2/4D_{\perp}$ and the system has modulated order.

The transition between the uniform and modulated orderings is abrupt for a one-component field and occurs along the line $c_{\perp} = -2\sqrt{-D_{\perp}\tilde{r}/5}$. For a two-component OP ($E_g$) we must also allow a relative phase between the two components of the field. In this case the uniform ordered phase is only stable for $c_{\perp} > 0$, and the modulated phase is now characterized by helical order with $\langle \eta(x) \rangle = \eta_* \{\cos(q_* x_3), \sin(q_* x_3)\}$. The uniform-modulated transition is now continuous. This does not reproduce the physics of HO, which has an abrupt transition between HO and AFM, and so we will henceforth neglect the possibility of a multicomponent order parameter. The schematic phase diagrams for this model are shown in Figure 1.

We will now proceed to derive the effective strain stiffness tensor $\lambda$ that results from the coupling of strain to the OP. The ultimate result, found in (17), is that $\lambda_X$
differs from its bare value \( C_X \) only for the symmetry \( X \) of the OP. Moreover, the effective strain stiffness does not vanish at the unordered–modulated transition, but exhibits a cusp. To show this, we will first compute the susceptibility of the OP, which will both be demonstrative of how the stiffness is calculated and prove useful in expressing the functional form of the stiffness. Then we will compute the strain stiffness using some tricks from functional calculus.

The susceptibility of a single component (\( B_{1g} \) or \( B_{2g} \)) OP to a thermodynamically conjugate field is given by

\[
\chi^{(-1)}(x, x') = \frac{\delta^2 F[\eta, \epsilon_\chi, \epsilon_\chi]}{\delta \eta(x) \delta \eta(x')} \bigg|_{\eta=\langle \eta \rangle} = \left[ \tilde{r} - c_{\parallel} \nabla_{\parallel}^2 \right. \\
\left. - c_{\perp} \nabla_{\perp}^2 + D_{\perp} q_{\perp}^4 + 12u(\eta(x))^2 \right] \delta(x - x'),
\]

where \( \{-1\} \) indicates a functional reciprocal in the sense that

\[
\int dx'' \chi^{(-1)}(x, x'') \chi(x'', x') = \delta(x - x').
\]

Taking the Fourier transform and integrating over \( q' \) we have

\[
\chi(q) = (\tilde{r} + c_0 q_{\parallel}^2 + c_{\perp} q_{\perp}^2 + D_{\perp} q_{\perp}^4 + 12u \sum_{q'} \langle \eta(q') \rangle \langle \eta(q-q') \rangle^{-1}. \tag{11}
\]

Near the unordered–modulated transition this yields

\[
\chi(q) = \frac{1}{c_{\parallel} q_{\parallel}^2 + D_{\parallel} (q_{\parallel}^2 - q_{\perp}^2)^2 + |\Delta \tilde{r}|} = \frac{1}{D_{\parallel} 1 + \xi_{\parallel} q_{\parallel}^2 + \xi_{\perp} (q_{\parallel}^2 - q_{\perp}^2)^2}, \tag{12}
\]

with \( \xi_{\parallel} = |\Delta \tilde{r}|/D_{\parallel} \) and \( \xi_{\perp} = (|\Delta \tilde{r}|/c_{\perp})^{-1/4} = \xi_{\perp} \xi_{\parallel} |t|^{-1/4} \). The reduced temperature and \( \xi_{\perp} = (D_{\perp}/aT_c)^{1/4} \) and \( \xi_{\parallel} = (c_0/aT_c)^{1/2} \) are the bare correlation lengths. Notice that the static susceptibility \( \chi(0) = (D_{\perp} q_{\perp}^4 + |\Delta \tilde{r}|)^{-1} \) does not diverge at the unordered–modulated transition. Though it anticipates a transition with Curie–Weiss-like divergence at \( \Delta \tilde{r} = -D_{\perp} q_{\perp}^4 \), this is cut off with a cusp at \( \Delta \tilde{r} = 0 \). We must emphasize that this is not the magnetic susceptibility because a \( B_{1g} \) or \( B_{2g} \) OP cannot couple linearly to a uniform magnetic field. The object defined in (9) is most readily interpreted as proportional to the two-point connected correlation function \( \langle \delta \eta(x) \delta \eta(x') \rangle = G(x, x') = k_B T \chi(x, x') \).

The strain stiffness is given in a similar way to the inverse susceptibility: we must trace over \( \eta \) and take the second variation of the resulting effective free energy functional of \( \epsilon \). Extremizing over \( \eta \) yields

\[
0 = \frac{\delta F[\eta, \epsilon]}{\delta \eta(x)} \bigg|_{\eta=\langle \eta \rangle} = \frac{\delta F_{\text{op}}[\eta]}{\delta \eta(x)} \bigg|_{\eta=\langle \eta \rangle} - b \epsilon_\chi(x), \tag{13}
\]

which explicitly gives \( \eta^*_0[\epsilon] \), the optimized OP conditioned on the strain. Since \( \eta^*_0 \) is a functional of \( \epsilon_\chi \) alone, only the stiffness \( \lambda_\chi \) can be modified from its bare value \( C_X \). Though this differential equation for \( \eta^*_0 \) cannot be solved explicitly, we can make use of the inverse function theorem. First, denote by \( \eta^{-1}_0[\eta] \) the inverse functional of \( \eta^*_0 \) implied by (13), which gives the function \( \epsilon_\chi \) corresponding to each solution of (13) it receives. This can immediately identify from (13) as \( \eta^{-1}_0[\eta](x) = b^{-1} (\delta F_{\text{op}}[\eta]/\delta \eta(x)) \). Now, we use the inverse function theorem to relate the functional reciprocal of the derivative of \( \eta^*_0[\epsilon] \) with respect to \( \epsilon_\chi \) to the derivative of \( \eta^{-1}_0[\eta] \) with respect to \( \eta_0 \), yielding

\[
\left( \frac{\delta \eta^*_0[\epsilon]}{\delta \epsilon_\chi(x')} \right)^{(-1)} = \frac{\delta \eta^{-1}_0[\eta](x)}{\delta \epsilon_\chi(x')} \bigg|_{\eta=\eta^*_0[\epsilon] = b^{-1} \frac{\delta F_{\text{op}}[\eta]}{\delta \eta(x)} \bigg|_{\eta=\eta^*_0[\epsilon]} \tag{14}
\]

Next, (13) and (14) can be used in concert with the ordinary rules of functional calculus to yield the second variation

\[
\frac{\delta^2 F[\eta^*_0[\epsilon], \epsilon]}{\delta \epsilon_\chi(x) \delta \epsilon_\chi(x')} = C_X \delta (x - x') - 2b \frac{\delta \eta^*_0[\epsilon]}{\delta \epsilon_\chi(x')} - b \int dx'' \frac{\delta^2 \eta^*_0[\epsilon]}{\delta \epsilon_\chi(x) \delta \epsilon_\chi(x'')} \epsilon_\chi(x''') + \int dx'' \frac{\delta^2 \eta^*_0[\epsilon]}{\delta \epsilon_\chi(x) \delta \epsilon_\chi(x'')} \delta \eta(x''') \bigg|_{\eta=\eta^*_0[\epsilon]} \tag{15}
\]
The strain stiffness is given by the second variation evaluated at the extremized strain \( \langle \epsilon \rangle \). To calculate it, note that evaluating the second variation of \( F_{\text{op}} \) in (14) at \( \langle \epsilon \rangle \) (or \( \eta_{\mu}(\langle \epsilon \rangle) = \langle \eta \rangle \)) yields
\[
\left( \frac{\delta \eta_{\mu}[\epsilon(x)]}{\delta \epsilon(x')} \right)_{x(\langle \epsilon \rangle)} = b^{-1} \chi^{-1}(x, x') + \frac{b}{C_{\chi}} \delta(x - x'),
\]
where \( \chi^{-1} \) is the op susceptibility given by (9). Upon substitution into (15) and taking the Fourier transform of the result, we finally arrive at
\[
\lambda_X(q) = C_X - b \left( \frac{1}{b \chi(q)} + \frac{b}{C_{\chi}} \right)^{-1} = C_X \left( 1 + \frac{b^2}{C_{\chi}} \chi(q) \right)^{-1}.
\]
(16)

Though not relevant here, this result generalizes to multicomponent ops. At \( q = 0 \), which is where the stiffness measurements used here were taken, this predicts a cusp in the static strain stiffness \( \lambda_X(0) \) of the form \( |\Delta \tilde{\epsilon}|^\gamma \) for \( \gamma = 1 \).

We have seen that the mean-field theory of a B\(_1g\) OP recreates the topology of the HO phase diagram and the temperature dependence of the B\(_1g\) strain stiffness at zero pressure. This theory has several other physical implications. First, the association of a modulated B\(_1g\) order with the HO phase implies a uniform B\(_{1g}\) order associated with the AFM phase, and moreover a uniform B\(_1g\) strain of magnitude \( \langle \epsilon_{B_{1g}} \rangle^2 = b^2/4 \bar{D}_L q^2 \), which corresponds to an orthorhombic phase. Orthorhombic symmetry breaking was recently detected in the AFM phase of UR\(_u\)Si\(_2\) using x-ray diffraction, a further consistency of this theory with the phenomenology of UR\(_u\)Si\(_2\) [22]. Second, as the Lifshitz point is approached from low pressure, this theory predicts that the modulation wavevector \( q_s \) should vanish continuously. Far from the Lifshitz point we expect the wavevector to lock into values commensurate with the space group of the lattice, and moreover that at zero pressure, where the RUS data here was collected, the half-wavelength of the modulation should be commensurate with the lattice spacing \( a_3 \approx 9.68 \text{ Å} \), or \( q_s = \pi/a_3 \approx 0.328 \text{ Å}^{-1} \) [29–32]. In between these two regimes, the ordering wavevector should shrink by jumping between ever-closer commensurate values in the style of the devil’s staircase [33]. This motivates future RUS experiments done at pressure, where the depth of the cusp in the B\(_1g\) stiffness should deepen (perhaps with these
commensurability jumps) at low pressure and approach zero like $q^4_\perp \sim (c_\perp/2D_\perp)^2$ near the Lifshitz point. The presence of spatial commensurability is not expected to modify the critical behavior otherwise [34].

There are two apparent discrepancies between the orthorhombic strain in the phase diagram presented by [22] and that predicted by our mean field theory. The first is the apparent onset of the orthorhombic phase in the HO state prior to the onset of AFM. As [22] notes, this could be due to the lack of an ambient pressure calibration for the lattice constant. The second discrepancy is the onset of orthorhombicity at higher temperatures than the onset of AFM. Susceptibility data sees no trace of another phase transition at these higher temperatures [35], and therefore we don’t expect there to be one. We do expect that this could be due to the high energy nature of x-rays as an experimental probe: orthorhombic fluctuations could appear at higher temperatures than the true onset of an orthorhombic phase.

Three dimensions is below the upper critical dimension $4_{\perp}/3$, and so mean field theory should break down sufficiently close to the critical point due to fluctuations, at the Ginzburg temperature [36, 37]. Magnetic phase transitions tend to have Ginzburg temperature of order one. The Ginzburg criterion is satisfied, $\eta$ is order one and the Landau–Ginzburg free energy expansion is no longer valid.

We have preformed a general treatment of phenomenological HO OPs with the potential for linear coupling to strain. The possibilities with consistent mean field phase diagrams are $B_{1g}$ and $B_{2g}$, and the only of these consistent with zero-pressure RUS data is $B_{1g}$, with a cusp appearing in the associated stiffness. In this picture, the HO phase is characterized by uniaxial modulated $B_{1g}$ order, while the AFM phase is characterized by uniform $B_{1g}$ order. The corresponding prediction of uniform $B_{1g}$ symmetry breaking in the AFM phase is consistent with recent diffraction experiments [22]. This work motivates both further theoretical work regarding a microscopic theory with modulated $B_{1g}$ order, and preforming RUS experiments at pressure that could further support or falsify this idea.

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