The analogy between the 'hidden order' and the orbital antiferromagnetism in $URu_{2-x}Fe_xSi_2$

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MATERIAL AND METHODS

Single crystals of Fe substituted URu_2Si_2 were grown in a tetra-arc furnace using the Czochralski method in an argon atmosphere. The quality of the synthesized single crystals was confirmed by x-ray diffraction measurements in a Bruker D8 Discover diffractometer.

The Raman spectra were acquired in a quasi-backscattering geometry along the crystallographic c-axis, using a continuous flow liquid helium optical cryostat. For excitation, the 752.5 nm (about 1.65 eV) Kr^+ ion laser line was used, with the spot size roughly $50\times100\,\mu m^2$. The power on the sample is about 12 mW for most temperatures, and kept below 6 mW to achieve the lowest temperatures. All temperatures shown were corrected for laser heating. The scattered light was analyzed and collected by a custom triple-grating spectrometer equipped with a liquid nitrogen cooled CCD detector. The intensities were corrected for the spectral response of the spectrometer and CCD.

We adopt Porto's notation to indicate the scattering geometries, where the directional vectors of incident and scattered light polarizations are denoted by $\mathbf{e}_i$ and $\mathbf{e}_s$, respectively. X=[100] and Y=[010] are aligned along crystallographic a-axis, X′=[110] and Y′=[1$\bar{1}$0] are aligned 45° to the a-axis, R=(X+iY)/$\sqrt{2}$ and L=(X-iY)/$\sqrt{2}$ are right and left circularly polarized, respectively. The measured spectral intensity in the $\mathbf{e}_i\mathbf{e}_s$ scattering geometry, $I_{\mathbf{e}_i\mathbf{e}_s}(\omega,T)$, is related to the Raman response function, $\chi''_{\mathbf{e}_i\mathbf{e}_s}(\omega,T) \equiv \mathrm{Im}[\chi_{\mathbf{e}_i\mathbf{e}_s}(\omega,T)]$, by the following: $I_{\mathbf{e}_i\mathbf{e}_s}(\omega,T) = [1+n(\omega,T)]\chi''_{\mathbf{e}_i\mathbf{e}_s}(\omega,T) + L(\omega,T)$, where $\chi_{\mathbf{e}_i\mathbf{e}_s}(\omega,T)$ is the Raman susceptibility, $n(\omega,T)$ is the Bose factor, and $L(\omega,T)$ is a background mainly resulting from luminescence of the sample. In the D_{4h} group, the Raman response in different scattering geometries are composed of the excitations from distinct symmetry channels as dictated by the Raman tensors [44]. The excitations accessible to Raman scattering from the ab-plane can be decomposed into 4 irreducible representations, i.e., A_{1g}, A_{2g}, B_{1g} and B_{2g}. However, we noticed that other than the phononic contribution, the response in B_{1g} and B_{2g} symmetry channels are the same and weakly temperature dependent. This suggests that electronic Raman contributions in the B_{1g} and B_{2g} channels are negligibly small, and the measured spectral intensity in RL scattering geometry, $I_{RL}(\omega,T) = [1+n(\omega,T)][\chi''_{B1g}(\omega,T)+\chi''_{B2g}(\omega,T)] + L(\omega,T)$, is predominantly contributed by luminescent background. Therefore, we subtract signal in RL scattering geometry as a background from all measured spectra presented in this paper.

GINZBURG-LANDAU FREE ENERGY

The coupling parameters in the Ginzburg-Landau free energy (Eq. 1) are chosen following the recipes given in Haule and Kotliar [20, 40] with slight adjustments to match the phase diagram in Fig. 1a, with all energies and temperatures given in the units of kelvins, and all pressure units are in GPa. The quadratic couplings are defined as $\alpha_{HO}(\omega_0, T, P) \equiv -\frac{1}{2}J_{HO}(\omega_0, P) + \tilde{\alpha}_{HO}(\omega_0, T, P)$ and $\alpha_{AF}(\omega_0, T, P) \equiv -\frac{1}{2}J_{AF}(\omega_0, P) + \tilde{\alpha}_{AF}(\omega_0, T, P)$, where $\omega_0 = 35\,\mathrm{K}$ is the effective energy separation between the singlet states in the minimal model, T is temperature, and P can be either hydrostatic pressure or effective chemical pressure. $J_{HO}(\omega_0, P) = \dfrac{\omega_0(1+a_1P)}{\tanh\left(\frac{\omega_0}{2T_{HO}}\right)}$ and $J_{AF}(\omega_0, P) = \dfrac{\omega_0(1+a_2P)}{\tanh\left(\frac{\omega_0}{2T_N}\right)}$ are the effective nearest neighbor exchange constants, with $T_{HO} = 17.5\,\mathrm{K}$ and $T_N = 15.5\,\mathrm{K}$. $\tilde{\alpha}_{HO}(\omega_0, T, P) = \frac{1}{2}\omega_0 \coth\left(\frac{\omega_0}{2T}\right)(1 - a_3P)$ and $\tilde{\alpha}_{AF}(\omega_0, T, P) = \frac{1}{2}\omega_0 \coth\left(\frac{\omega_0}{2T}\right)(1 + a_3P)$ are the effective on-site couplings. $\beta(\omega_0, P) = \dfrac{\omega_0\left[\sinh\left(\frac{\omega_0}{T}\right) - \frac{\omega_0}{T}\right]\cosh^2\left(\frac{\omega_0}{2T}\right)}{8\sinh^4\left(\frac{\omega_0}{2T}\right)}$ is the quartic coupling.

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