Chirality density wave of the 'hidden order' phase in URu_2Si_2

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H.-H. Kung, R. E. Baumbach, E. D. Bauer, V. K. Thorsmølle, W.-L. Zhang, K. Haule, J. A. Mydosh & G. Blumberg

A second-order phase transition is associated with emergence of an "order parameter" and a spontaneous symmetry breaking. For the heavy fermion superconductor URu_2Si_2, the symmetry of the order parameter associated with its ordered phase below 17.5 K has remained ambiguous despite 30 years of research, and hence is called "hidden order" (HO). Here we use polarization resolved Raman spectroscopy to specify the symmetry of the low energy excitations above and below the HO transition. These excitations involve transitions between interacting heavy uranium $5f$ orbitals, responsible for the broken symmetry in the HO phase. From the symmetry analysis of the collective mode, we determine that the HO parameter breaks local vertical and diagonal reflection symmetries at the uranium sites, resulting in crystal field states with distinct chiral properties, which order to a commensurate chirality density wave ground state.

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- Ionic U can adopt six different valences, usually U^{4+} with two $5f$ electrons

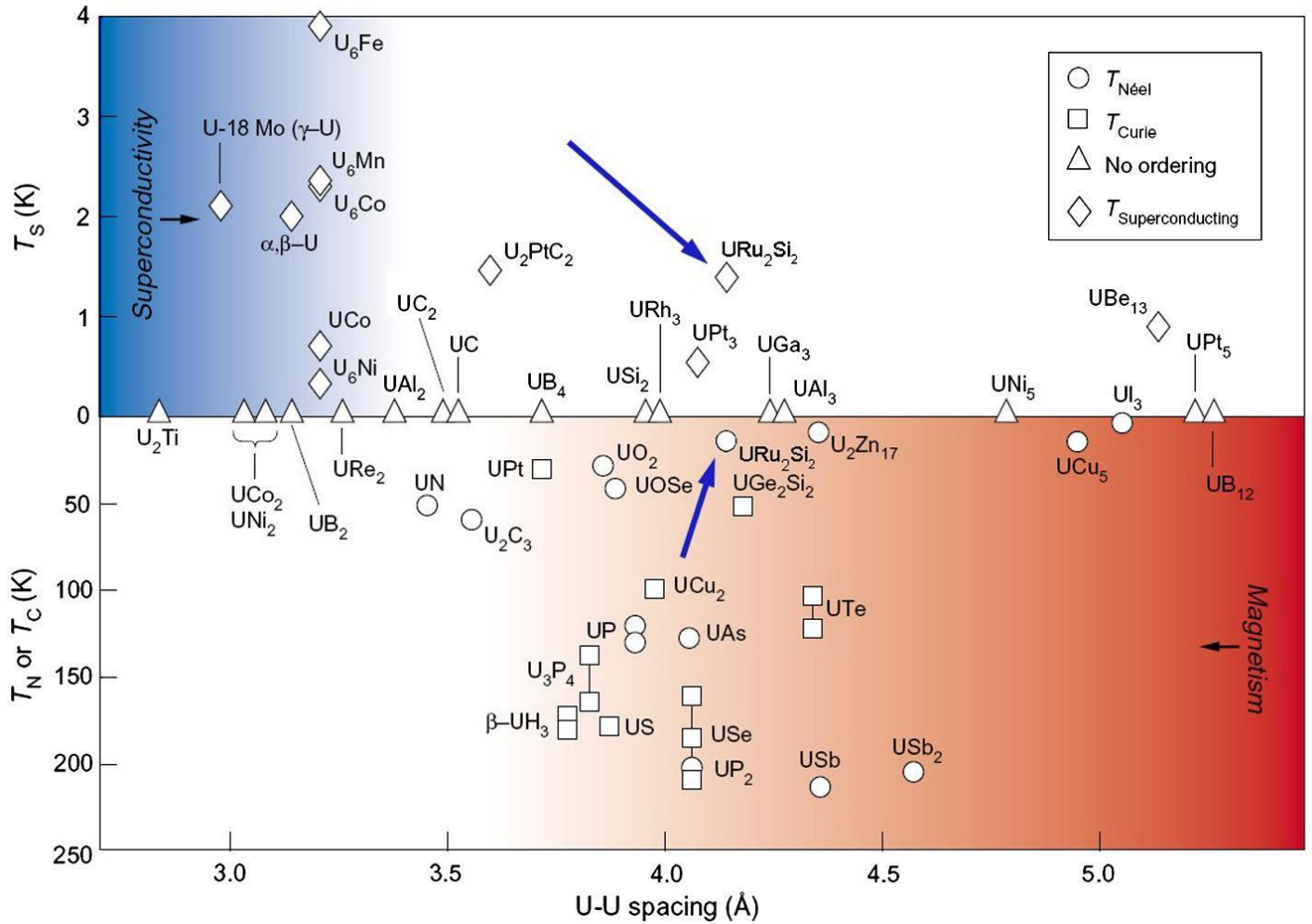

Figure : The Hill plot for various U based intermetallic compounds. Bottom arrow indicates hidden-order (HO) transition temperature T_0 and the top arrow indicates its superconducting transition temperature (Hill, 1970).

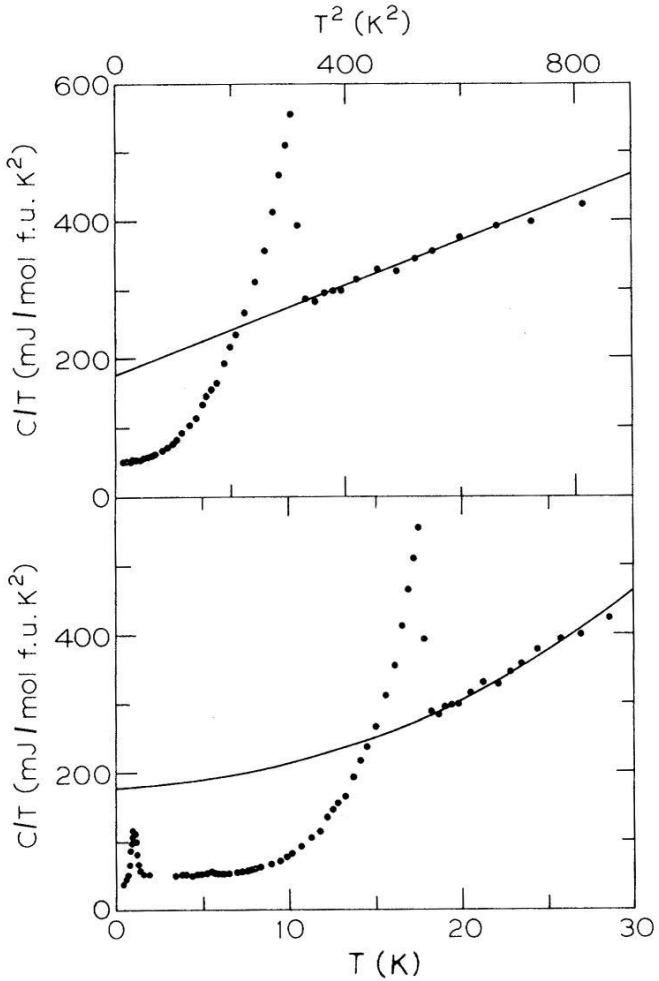

Figure : Electronic specific heat as a function of temperature for URu_2Si_2 (Palstra et al., PRL 1985)

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- A: commensurate chirality density wave

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- Presence of $e - e$ Coulomb interactions and electron spin orbit interactions considerably lift the degeneracy as revealed by atomic spectroscopies

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- The first two rules only determine the values of L and S, what about total angular momentum $J = L + S$, which can take all integral values between $|L - S|$ and $L + S$

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- Nine fold degeneracy can be labeled using $J_z \in \{-4, 4\}$

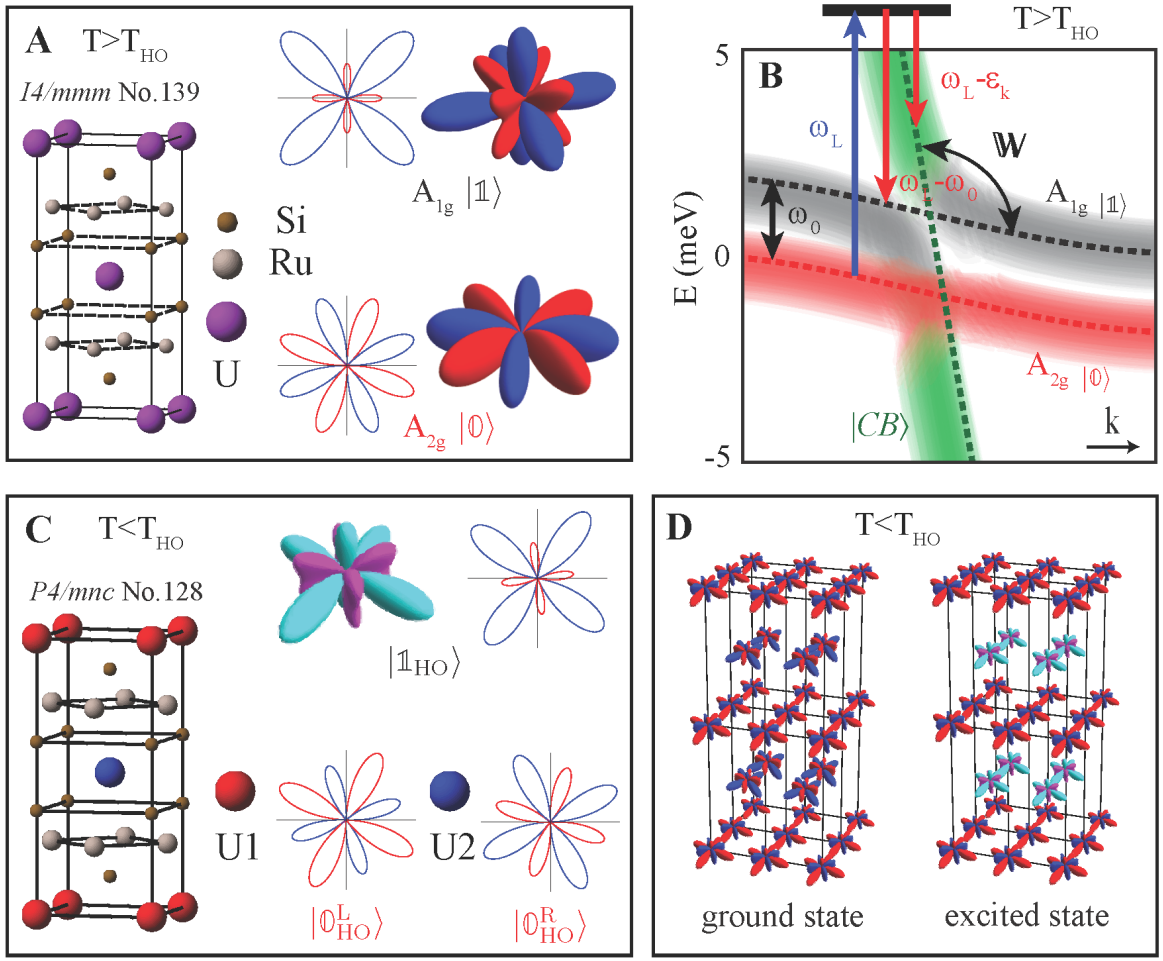

Figure : Schematics of the local symmetry and band structure of the quasi-localized states in the minimal model, above and below T_{HO}.(**A**) The crystal structure of URu_2Si_2 in the paramagnetic phase. Presented in 3D and *xy*-plane cut are wave functions that show the symmetry of the A_{2g} state $|\mathbb{0}\rangle$ and A_{1g} state $|\mathbb{1}\rangle$, where the positive (negative) amplitude is denoted by red (blue) color. The A_{1g} state is symmetric with respect to the vertical and diagonal reflections, while the A_{2g} state is antisymmetric with respect to these reflections.

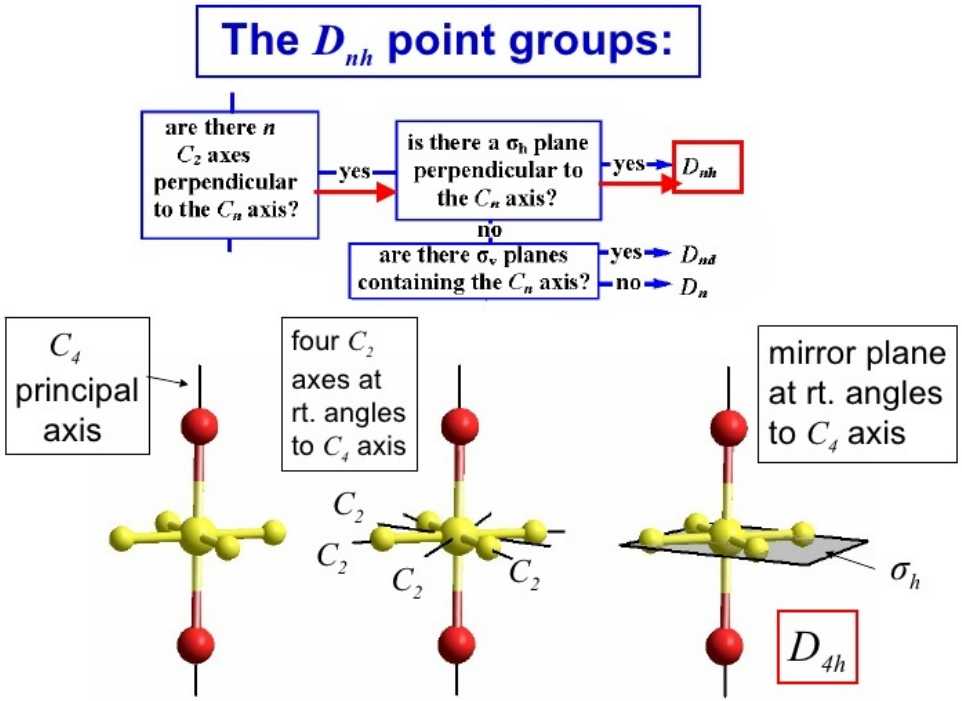

Figure : URu_2Si_2 crystallizes in a body-centered tetragonal structure belonging to the $\mathbb{D}_{4\mathbb{h}}$ point group.

Table S1: The crystal field states of U-5f electrons with two electrons in the $j = 5/2$ subshell, categorized by irreducible representations (IRR) of the $\mathbb{D}_{4h}$ group. These states have total angular moment $J = 4$ and magnetic quantum number M_j runs from $-4 \cdots 4$. The states on the right hand site are labeled by their M_j value, e.g. $|0\rangle \equiv |J = 4, M_j = 0\rangle$

.

IRR	state
$A_{2g}\,(\Gamma_2)$	$\|0\rangle = \frac{i}{\sqrt{2}}(\|4\rangle - \|-4\rangle)$
$A_{1g}\,(\Gamma_1^{(1)})$	$\|1\rangle = \frac{\cos\theta}{\sqrt{2}}(\|4\rangle + \|-4\rangle) - \sin\theta\,\|0\rangle$
$A_{1g}\,(\Gamma_1^{(2)})$	$\|2\rangle = \frac{\sin\theta}{\sqrt{2}}(\|4\rangle + \|-4\rangle) + \cos\theta\,\|0\rangle$
$E_g\,(\Gamma_{5,1}^{(1)})$	$\|3\rangle = \cos\phi\,\|-3\rangle + \sin\phi\,\|1\rangle$
$E_g\,(\Gamma_{5,2}^{(1)})$	$\|4\rangle = \cos\phi\,\|3\rangle + \sin\phi\,\|-1\rangle$
$E_g\,(\Gamma_{5,1}^{(2)})$	$\|5\rangle = \sin\phi\,\|-3\rangle - \cos\phi\,\|1\rangle$
$E_g\,(\Gamma_{5,2}^{(2)})$	$\|6\rangle = \sin\phi\,\|3\rangle - \cos\phi\,\|-1\rangle$
$B_{1g}\,(\Gamma_3)$	$\|7\rangle = \frac{1}{\sqrt{2}}(\|2\rangle + \|-2\rangle)$
$B_{2g}\,(\Gamma_4)$	$\|8\rangle = \frac{i}{\sqrt{2}}(\|2\rangle - \|-2\rangle)$

Figure : In the crystal environment of URu_2Si_2, the nine states split into seven energy levels denoted by irreducible representations of the $\mathbb{D}_{4h}$ group: 5 singlet states $2A_{1g} \oplus A_{2g} \oplus B_{1g} \oplus B_{2g}$ and 2 doublet states $2E_g$.

Mulliken Labels for Representations

- **Non-Degenerate Representations = A or B**

  Character under identity E = 1; (symmetric)
  Character under Principal axis = 1 then Symbol = A

  Character under identity E = 1; (symmetric)
  Character under Principal axis = -1 (anti-symmetric)
  Symbol = B

When symmetric to inversion (i): add g = gerade (e.g. A_{1g})
When anti-symmetric to inversion: add u = ungerade (A_{1u})

- **Degenerate Representations:**

  Doublet: Symbol =E (Example: E_g in O_h point group)
  Triplet: Symbol = T (Example: T_{2g} in O_h point group)

Figure : Low energy minimal model including lowest three states only have A_{1g} and A_{2g} states

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- They perform Raman in various geometries. The six proper scattering geometries are denoted as $\mathbf{e}_s\mathbf{e}_i$ =XX, XY, X′X′, X′Y′, RR and RL, with $\mathbf{e}_i$ being the direction vector for incident light polarization, and $\mathbf{e}_s$ being the scattered light polarization. X=[100], Y=[010] are aligned along crystallographic axes, X′=[110], Y′=[1$\bar{1}$0] are aligned 45° to the *a*-axes, R=(X+*i*Y)/$\sqrt{2}$ and L=(X-*i*Y)/$\sqrt{2}$ are right and left circularly polarized light

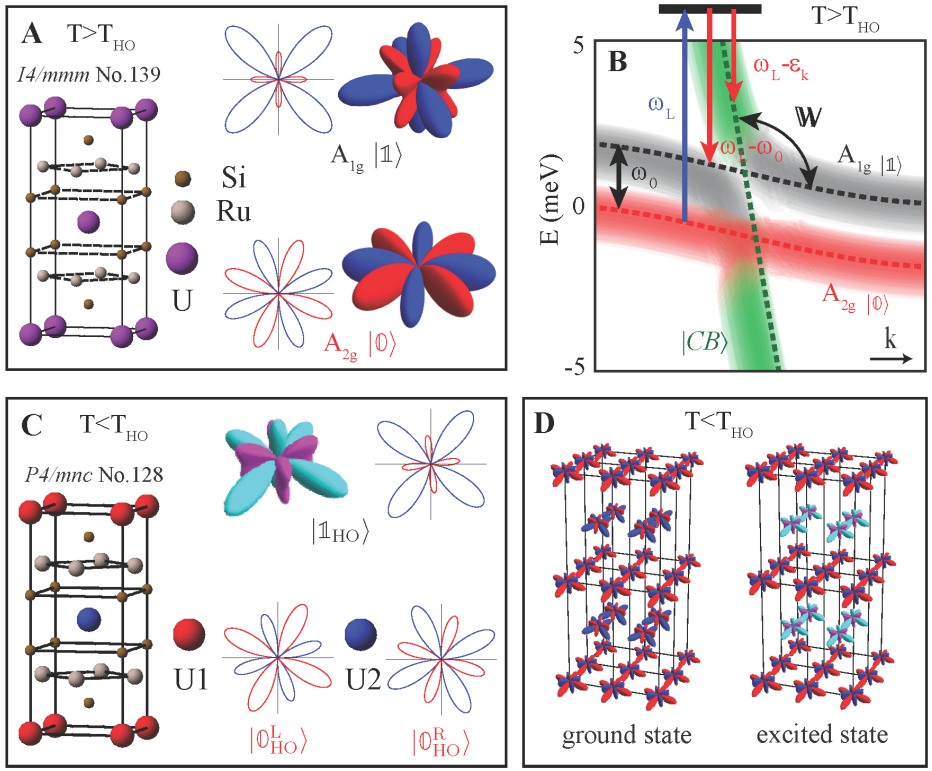

Figure : (**B**) Schematic of the band structure of a minimal model in the paramagnetic state. The green dashed line denotes the conduction band $|CB\rangle$, the red and black dashed lines denote crystal field states of the U *5f* electrons: the ground state $|\mathbb{0}\rangle$ and the first excited state $|\mathbb{1}\rangle$. A cartoon of the Raman process is shown, where the blue and red arrows denote the incident and scattered light, respectively. ω_L is the incoming photon energy (not in scale), $\mathbb{W}$ is the hybridization strength between $|\mathbb{1}\rangle$ and $|CB\rangle$, ω_0 and ε_k are the resonance energies for $|\mathbb{0}\rangle \rightarrow |\mathbb{1}\rangle$ and $|\mathbb{0}\rangle \rightarrow |CB\rangle$, respectively.

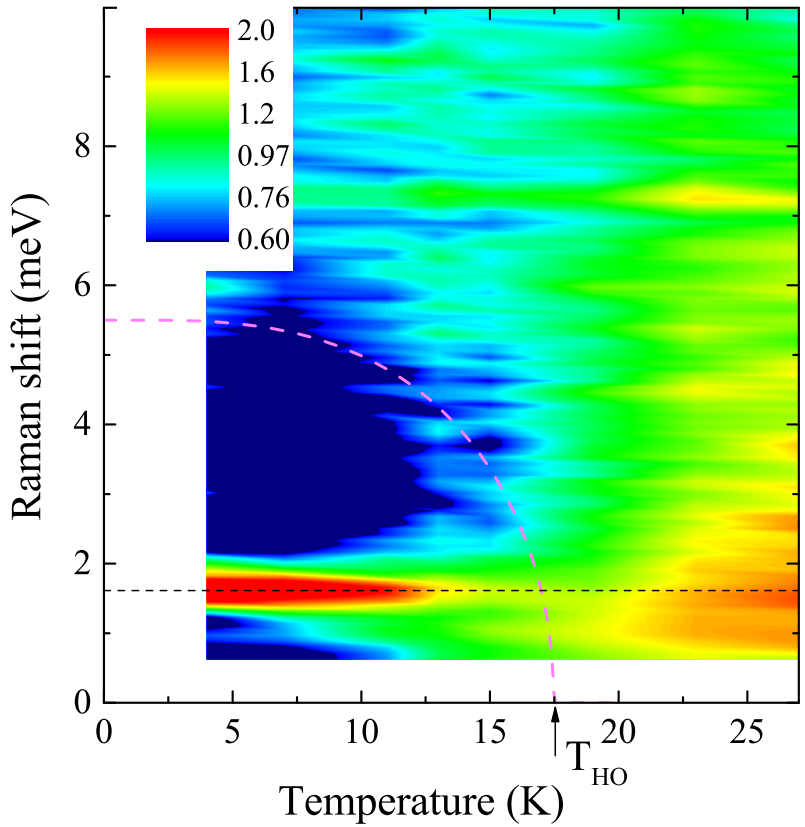

Figure : Raman response in the XY scattering geometry as function of temperature and Raman shift energy. The contour plot shows the temperature evolution of the low energy Raman response in the XY scattering geometry. A sharp excitation at 1.6 meV (indicated by the black dashed line) emerges below T_{HO}. The mode's full width at half maximum decreases on cooling to about 0.3 meV at 4 K. A gap-like suppression develops to a magnitude of about 6 meV at 4 K. The pink dashed line shows the temperature dependence of a gap expected from a mean-field BCS model with a transition temperature of 17.5 K.

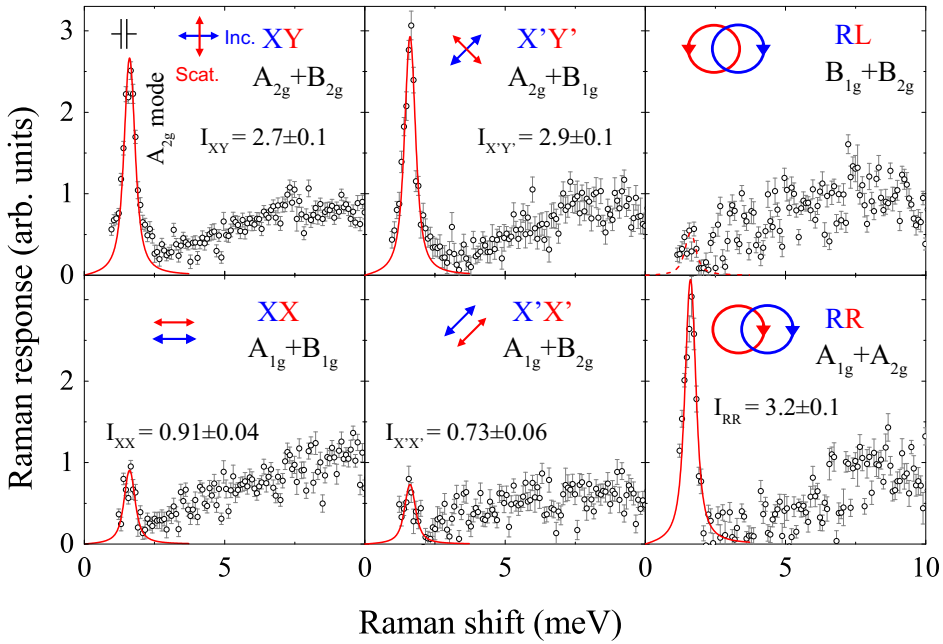

Figure : The Raman response in six proper scattering geometries at 7 K. The data are shown in black circles, where the error bars show one standard deviation. The red solid lines are fits of the in-gap mode to a Lorentzian, and the fitted intensity using the method of maximum likelihood is shown in each panel. By decomposition, the in-gap mode intensity in each symmetry channels are: $I_{A_{2g}} = 2.6 \pm 0.1$, $I_{A_{1g}} = 0.7 \pm 0.1$, $I_{B_{1g}} = 0.3 \pm 0.1$, and $I_{B_{2g}} = 0.1 \pm 0.1$. The full width at half maximum of the in-gap mode is about 0.5 meV at 7 K (deconvoluted with instrumental resolution of 0.15 meV, shown in the XY panel)

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- The in-gap mode intensity in the A_{1g} channel is about four times weaker than in the A_{2g} channel.

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- This signifies the breaking of the local vertical and diagonal reflection symmetries at the uranium sites in the HO phase.
- Similarly, the tiny intensity leakage into the RL scattering geometry measure the strength of orthorhombic distortion due to broken four-fold rotational symmetry.

- When the reflection symmetries are broken, an A_{2g}-like interaction operator $\Psi_{HO} \equiv V \left|\mathbb{1}\right\rangle \left\langle\mathbb{0}\right|$ mixes the $\left|\mathbb{0}\right\rangle$ and $\left|\mathbb{1}\right\rangle$ states leading to two new local states:
  $\left|\mathbb{0}^{L}_{HO}\right\rangle \approx (1 - \frac{V^2}{2\omega_0^2}) \left|\mathbb{0}\right\rangle - \frac{V}{\omega_0} \left|\mathbb{1}\right\rangle$ and
  $\left|\mathbb{0}^{R}_{HO}\right\rangle \approx (1 - \frac{V^2}{2\omega_0^2}) \left|\mathbb{0}\right\rangle + \frac{V}{\omega_0} \left|\mathbb{1}\right\rangle$, with V being the interaction strength.

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- The choice of either the left-handed or the right-handed state on a given uranium site, $\left|\mathbb{0}^{L}_{HO}\right\rangle$ or $\left|\mathbb{0}^{R}_{HO}\right\rangle$, defines the local chirality in the HO phase

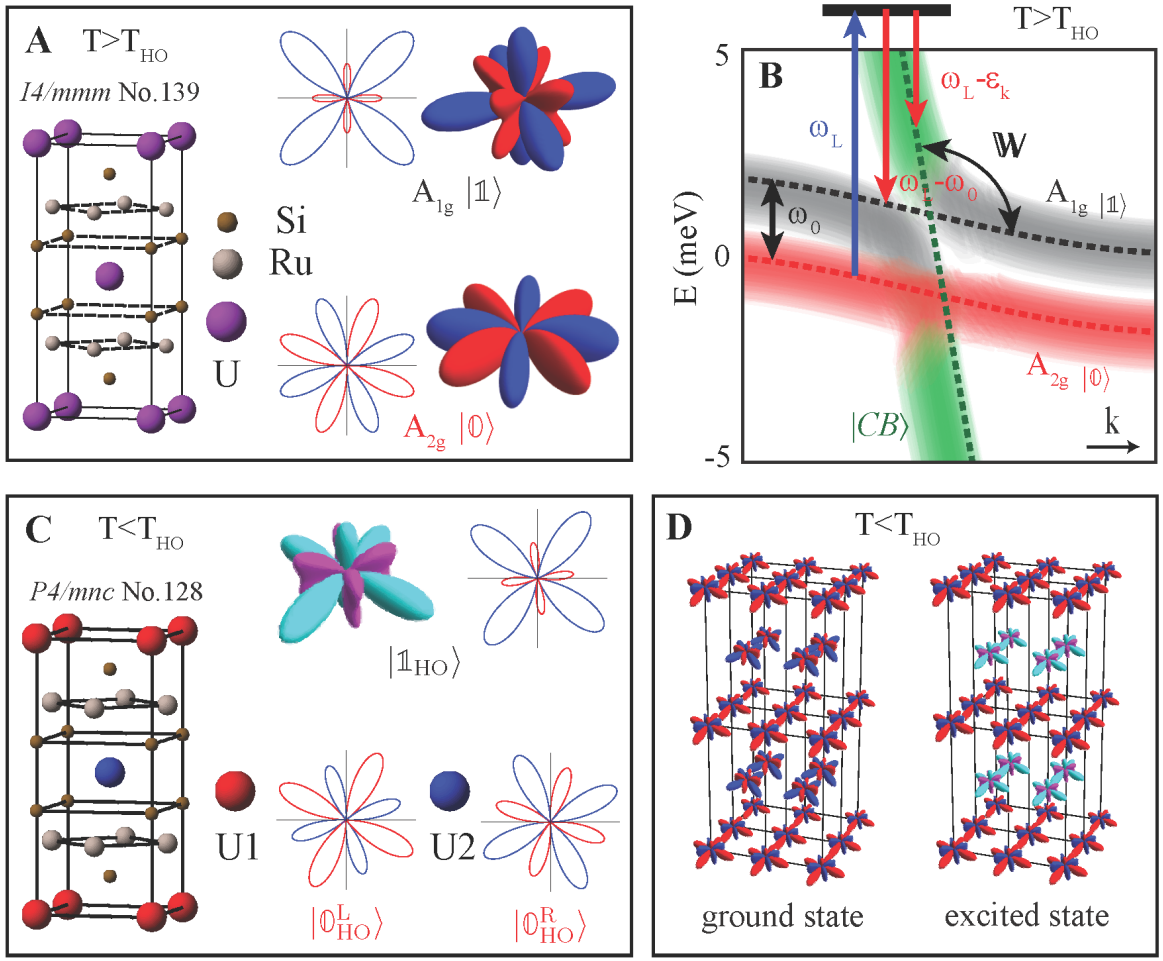

Figure : (**D**) Show schematics of chirality density wave, where the chiral states are staggered in the lattice (left). The right figure shows one of the possible excited state of the chirality density wave, where the chiral state $|\mathbb{O}^{R}_{HO}\rangle$ at lattice site U2 is excited to $|\mathbb{1}_{HO}\rangle$.

- A local order parameter of primary A_{2g} symmetry, breaking vertical/diagonal reflections, with subdominant B_{1g} component, breaking four-fold rotational symmetry, can be expressed in terms of the composite hexadecapole local order parameter of the form:
$\pm V[(J_x - J_y)(J_x + J_y)(J_x J_y + J_y J_x) + (J_x J_y + J_y J_x)(J_x + J_y)(J_x - J_y)]$

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- where J_x, J_y are in-plane angular momentum operators
- A spatial order alternating the sign of this hexadecapole for neighboring basal planes is the chirality density wave that consistently explains the HO phenomena as it is observed by Raman and neutron scattering.

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- Thank you for your attention !!