





Spin, charge and energy transport in novel materials Hvar, Croatia, October 1 - 7, 2017

Challenges and Implications of Hidden Order in URu₂Si₂



I. The Mystery

P. Chandra Rutgers

- II. The Story So Far
- III. A New Type of Order Parameter?













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Challenges and Implications of Hidden Order in URu₂Si₂



Jennifer Trinh (UC Santa Cruz) Arthur P. Ramirez (UC Santa Cruz)

Rebecca Flint (Iowa State) Piers Coleman (Rutgers)





P. Chandra

Rutgers





The Mystery of URu₂Si₂

$$\Delta S = \int_0^{T_0} \frac{C_V}{T} dT$$

=0.14 x 17.5 K =2.45 J/mol/K =**0.42 R In 2**

Large Order Parameter Expected

"Textbook" Ordering Transition at T = 17.5 K

Precursor to Superconductivity at T = 1.2 K



Yet....

Order parameter undetected after more than 25 years of research.



The Mystery of URu₂Si₂

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=0.42 R ln 2

What is the nature of the quasiparticle excitations

and

the Broken Symmetries

associated with this

Hidden Order Phase ??





Hidden Order: State of Matter where the Correlations Cannot be Identified

New Forms of Order



Dana and Omnes (1923)

Hidden Order: State of Matter where the Correlations Cannot be Identified

Competing/Intertwined Order



Forms of Order Beyond the Landau Classification ??

Topological Order



Forms of Order Beyond the Landau Classification ??

New Types of Symmetry-Breaking Order Parameters ??



Can order parameters, like excitations, fractionalize ??

$$\psi = \sqrt{\text{Multipole}} = \text{Spinorial OP}$$

PC, P. Coleman, R. Flint (2013).









The Mystery of URu₂Si₂

 $\Delta S = \int_{0}^{T_{0}} \frac{C_{V}}{T} dT = 0.14 \text{ x } 17.5 \text{ K} \\ = 2.45 \text{ J/mol/K}$

=0.42 R ln 2

Large entropy of condensation.





Broken Symmetry: ?? Order Parameter : ??

What is the nature of the hidden order?





What is the nature of the hidden order?

25 Years of Theoretical Proposals

Landau Theory Shah et al. ('00) "Hidden Order" Ramirez et al, '92 (Quadrupolar SDW) Ikeda and Ohashi '98 (d-density wave) Okuno and Miyake '98 (composite) Tripathi, Coleman, Mydosh and PC, '02 (orbital afm) Dori and Maki, '03 (Unconventional SDW) Mineev and Zhitomirsky, '04 (SDW) Varma and Zhu, '05 (Spin-nematic) Ezgar et al '06 (Dynamic symmetry breaking) Fujimoto, '11 (Spin-nematic) Ikeda et al '12 (Rank 5 nematic) Tanmoy Das '12 (Topological Spin-nematic) Barzykin & Gorkov, '93 (three-spin correlation) Santini & Amoretti, '94, Santini ('98) (Quadrupole order) Amitsuka & Sakihabara (⁵, Quadrupolar doublet, '94) Kasuya, '97 (U dimerization) Kiss and Fazekas '04, (Rank 3 octupolar order) Haule and Kotliar '09 (Rank 4 hexa-decapolar) Rau and Kee '12 (Rank 5 pseudo-spin vector) Pepin et al '10 (Spin liquid/Kondo Lattice) Dubi and Balatsky, '10 (Hybridization density wave)

Kondo Lattice

Itinerant

Local

Importance of Ising Anisotropy to HO Problem



U moments are Ising $\langle +|J_{\pm}|angle=0$ Integer S ($5f^2$)

Importance of Ising Anisotropy to HO Problem



Non-spinflip ($\Delta J_z = 0$) magnetic excitations also have Ising character !!

Inelastic Neutrons (Broholm et al, 91) Raman (Buhot et al, Kung et al., 15)





$$M \propto \cos \left[2\pi \frac{\text{Zeeman}}{\text{cyclotron}} \right]$$









$$\frac{m^*}{m_e}g(\theta) = 2n+1$$





M. M. Altarawneh, N. Harrison, S. E. Sebastian, et al., PRL (2011). H. Ohkuni *et al.*, Phil. Mag. B 79, 1045 (1999).

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$$\frac{g_c}{g_{\perp}} \ge 30 \quad \rightarrow \quad \frac{\chi_c^P}{\chi_{\perp}^P} \sim \left(\frac{g_c}{g_{\perp}}\right)^2 > 900$$

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$$\frac{m^*}{m_e}g(\theta) = 2n+1$$



Quasiparticle with giant Ising anisotropy > 30. Pauli susceptibility anisotropy > 900

 $\langle \mathbf{k}\sigma | J_{\pm} | \mathbf{k}\sigma' \rangle = 0$



Ising QP's pair condense.

Quasiparticle with giant Ising anisotropy > 30. Pauli susceptibility anisotropy > 900

 $\langle \mathbf{k}\sigma | J_{\pm} | \mathbf{k}\sigma' \rangle = 0$



Ising 5f doublet degenerate to within $2\Delta\sim 5K$



Ising QP's pair condense.

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Ising 5f doublet degenerate to within $2\Delta\sim 5K$





Ising QP's pair condense.

Quasiparticle with giant Ising anisotropy > 30. Pauli susceptibility anisotropy > 900

Electrons hybridize with Ising 5f state to form Landau quasiparticles. $\langle {f k}\sigma|J_\pm|{f k}\sigma'
angle=0$



.. Integer spin M



 $|\Gamma,\pm\rangle = a|\pm 3\rangle + b|\mp 1\rangle$ "\Gamma_5" non-Kramers doublet 5f²

.. Integer spin M



 $|\Gamma,\pm\rangle = a|\pm 3\rangle + b|\mp 1\rangle$ "\Gamma_5" non-Kramers doublet 5f²

... Integer spin M



"Γ₅" non-Kramers doublet 5f²



```
|\Gamma,\pm\rangle = a|\pm 3\rangle + b|\mp 1\rangle
"\Gamma_5" non-Kramers doublet 5f<sup>2</sup>
```



 $|\Gamma,\pm\rangle = a|\pm 3\rangle + b|\mp 1\rangle$ "\Gamma_5" non-Kramers doublet 5f² Hybridization is a spinor.


 $|\Gamma,\pm\rangle = a|\pm 3\rangle + b|\mp 1\rangle$ "\Gamma_5" non-Kramers doublet 5f²





Kramers index K: quantum no of *double* time reversal $\theta x \theta = \theta^2$.

$$\Theta^2 |\psi\rangle = K |\psi\rangle$$



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Half-integer spins change sign, integer spins do not.

 $K = (-1)^{2J}$



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Since the microscopic Hamiltonian must be Kramers-invariant,



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 $K=(-1)^{2J}$ Since the microscopic Hamiltonian must be Kramers-invariant, $V=-V^{2\pi}$



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$$\Theta^2 |\psi\rangle = K |\psi\rangle = |\psi^{2\pi}\rangle$$

Half-integer spins change sign, integer spins do not.

Hybridization transforms as a 1/2 integer spin.

 $K = (-1)^{2J}$ Since the microscopic Hamiltonian must be Kramers-invariant, V

 $V \equiv -V$



Kramers index K: quantum no of *double* time reversal $\theta x \theta = \theta^2$.

 $\theta^2 = 2\pi$ rotation:

$$\Theta^2 |\psi\rangle = K |\psi\rangle = |\psi^{2\pi}\rangle$$

Half-integer spins change sign, integer spins do not.

 $K = (-1)^{2J}$ Since the microscopic Hamiltonian must be Kramers-invariant,

 $V = -V^{2\pi}$

Hybridization transforms as a 1/2 integer spin. Unlike magnetism, it breaks **double time reversal. A new kind of order parameter.**

Measuring the hybridization gap





Hybridization is the Order Parameter !!??!!

Morr et al. (10) Dubi and Balatsky (10)

In conventional heavy fermion materials a hybridization derives from virtual excitations between a Kramers doublet and an excited singlet. A uniform hybridization breaks no symmetry and develops as a cross-over.







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H. Amitsuka and T. Sakakibara, J. Phys. Soc. Japan 63, 736-47 (1994).

But if the ground-state is a non-Kramer's doublet, the Kondo effect occurs via an *excited Kramer's doublet*.



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non-
Kramers
$$\Gamma_5$$
 == $|5f^2, \alpha\rangle = \hat{\chi}^{\dagger}_{\alpha}|0\rangle$
(K=+1)

H. Amitsuka and T. Sakakibara, J. Phys. Soc. Japan 63, 736-47 (1994).

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Kramers
(K=-1)
$$\Gamma_7 = [5f^3, \sigma\rangle = \hat{\Psi}^{\dagger}_{\sigma}|0\rangle$$

non-
Kramers
(K=+1) $\Gamma_5 = [5f^2, \alpha\rangle = \hat{\chi}^{\dagger}_{\alpha}|0\rangle$

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Kramers Γ_5 $= |5f^2, \alpha\rangle = \hat{\chi}^{\dagger}_{\alpha} |0\rangle$ (K=+1)

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$$5f^3,\sigma\rangle\langle 5f^2,\alpha|=\hat{\Psi}^{\dagger}_{\sigma}\hat{\chi}_{\alpha}$$



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 $|5f^3,\sigma\rangle\langle 5f^2,\alpha|=\hat{\Psi}^{\dagger}_{\sigma}\hat{\chi}_{\alpha}$





$$5f^3,\sigma\rangle\langle 5f^2,\alpha|=\hat{\Psi}^{\dagger}_{\sigma}\hat{\chi}_{\alpha}$$





$$|5f^3,\sigma\rangle\langle 5f^2,\alpha|\longrightarrow \langle\hat{\Psi}^{\dagger}_{\sigma}\rangle\hat{\chi}_{\alpha}$$



("Magnetic Higgs Boson")

$$\Psi = \begin{pmatrix} \langle \Psi_{\uparrow} \rangle \\ \langle \Psi_{\downarrow} \rangle \end{pmatrix}$$

$$|5f^3,\sigma\rangle\langle 5f^2,\alpha|\longrightarrow \langle\hat{\Psi}^{\dagger}_{\sigma}\rangle\hat{\chi}_{\alpha}$$



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Quasiparticles acquire the Ising anisotropy of the non-Kramers doublet.



("Magnetic Higgs Boson")

$$\Psi = \begin{pmatrix} \langle \Psi_{\uparrow} \rangle \\ \langle \Psi_{\downarrow} \rangle \end{pmatrix}$$

Landau Theory of Hastatic Order

$$\Psi = \begin{pmatrix} \Psi_{\uparrow} \\ \Psi_{\downarrow} \end{pmatrix} \qquad f[T, P] = \alpha (T_c - T) |\Psi|^2 + \beta |\Psi|^4 - \gamma (\Psi^{\dagger} \sigma_z \Psi)^2$$
$$\gamma = \delta (P - P_c)$$



AFM: P>Pc

$$\Psi_A \sim \begin{pmatrix} 1 \\ 0 \end{pmatrix}, \Psi_B \sim \begin{pmatrix} 0 \\ 1 \end{pmatrix}$$

Large f-moment

HO: P<Pc

$$\Psi_A \sim \frac{1}{\sqrt{2}} \begin{pmatrix} e^{-i\phi/2} \\ e^{i\phi/2} \end{pmatrix}, \Psi_B \sim \frac{1}{\sqrt{2}} \begin{pmatrix} -e^{-i\phi/2} \\ e^{i\phi/2} \end{pmatrix}$$

No f-moment: large Ising fluctuations

Landau Theory of Hastatic Order

$$\Psi = \begin{pmatrix} \Psi_{\uparrow} \\ \Psi_{\downarrow} \end{pmatrix} \qquad f[T, P] = \alpha (T_c - T) |\Psi|^2 + \beta |\Psi|^4 - \gamma (\Psi^{\dagger} \sigma_z \Psi)^2$$
$$\gamma = \delta (P - P_c)$$



Spin Flop-like Transition between HO and AFM₂₇

$$|\Gamma_{7}^{+},\sigma\rangle \equiv \Psi_{\sigma}^{\dagger}|0\rangle = \uparrow$$
$$E_{b}$$
$$|\pm\rangle \equiv \chi_{\pm}^{\dagger}|0\rangle = \downarrow$$

$$5f^{2} \rightleftharpoons 5f^{1} + e^{-} \psi^{\dagger}_{\Gamma\sigma}(j) = \sum_{\mathbf{k}} \left[\Phi^{\dagger}_{\Gamma}(\mathbf{k}) \right]_{\sigma\tau} c^{\dagger}_{\mathbf{k}\tau} e^{-i\mathbf{k}\cdot\mathbf{R}_{j}}$$
$$H_{VF}(j) = V_{6} \psi^{\dagger}_{\Gamma_{6}\pm}(j) |\Gamma_{7}^{+}\pm\rangle \langle \Gamma_{5}\pm| + V_{7} \psi^{\dagger}_{\Gamma_{7}\mp}(j) |\Gamma_{7}^{+}\mp\rangle \langle \Gamma_{5}\pm| + \mathrm{H.c.}$$

$$|\Gamma_{7}^{+},\sigma\rangle \equiv \Psi_{\sigma}^{\dagger}|0\rangle = \uparrow$$
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$$H_{VF}(j) = V_{6} \psi^{\dagger}_{\Gamma_{6}\pm}(j) \Psi^{\dagger}_{j\pm} \chi_{j\pm} + V_{7} \psi^{\dagger}_{\Gamma_{7}\mp}(j) \Psi^{\dagger}_{j\mp} \chi_{j\pm} + \text{H.c.}$$

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$$\langle \Psi_{j}^{\dagger} \rangle = |\Psi| \begin{pmatrix} \mathrm{e}^{i(\mathbf{Q} \cdot \mathbf{R}_{j} + \phi)/2} \\ \mathrm{e}^{-i(\mathbf{Q} \cdot \mathbf{R}_{j} + \phi)/2} \end{pmatrix}, \qquad (\phi = \pi/4).$$

$$|\Gamma_{7}^{+},\sigma\rangle \equiv \Psi_{\sigma}^{\dagger}|0\rangle = \uparrow$$
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$$H_{VF} = \sum_{\mathbf{k}} c_{\mathbf{k}}^{\dagger} \mathcal{V}_{6}(\mathbf{k}) \chi_{\mathbf{k}} + c_{\mathbf{k}}^{\dagger} \mathcal{V}_{7}(\mathbf{k}) \chi_{\mathbf{k}+\mathbf{Q}} + \text{h.c.}$$

Superconductivity: Giant Ising Anisotropy



Ising QP's pair condense.

θ

0

 $g(\theta) \propto \cos(\theta)$

Does this behavior survive to higher temperatures in the Hidden Order phase ??

The Nonlinear Susceptibility in a Tetragonal Environment

Field-Dependent Part of Free Energy

$$F = -\chi_1(\theta) \frac{H^2}{2} - \chi_3(\theta, \phi) \frac{H^4}{4}$$



 $\Delta \chi_3(\theta)$ is a direct thermodynamic probe of $g(\theta)$ at the Hidden Order transition !!


$$\chi_1(\theta, T) = \chi_1^{(0)} + \chi_1^{Ising}(T)\cos^2\theta$$



 $\Delta \chi_3(\theta) = \Delta \chi_3^{Ising}(T) \cos^4 \theta$

Robustness of Ising Anisotropy



Ising Quasiparticles at the Hidden Order Transition !!

$$F[\vec{H}] = F[H_z] \to H_{Zeeman} \propto -J_z B_z$$

Single-Ion Physics

U moments are Ising $\langle +|J_{\pm}|-\rangle = 0$ Integer S ($5f^2$) Confirmed by DMFT and high-resolution RIXS (Haule, Kotliar 09) (Wray et al. 15)

Also underlying itinerant ordering process (with Ising anisotropy!)



How to reconcile single-ion and itinerant perspectives ??





P. Coleman, R. Flint and PC, Nature 49

Nature 493, 611 (2013) Phil. Mag. 94, 32–33 (2014) 35 PRB 91, 205103 (2015)







P. Coleman, R. Flint and PC, Nature 493, 611 (2013)

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P. Coleman, R. Flint and PC, Nature 493, 611 (2013)

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Coherent Admixture of Spin 1/2 Conductio Electrons with Integer Spin f-states.



P. Coleman, R. Flint and PC, Nature 493, 611 (2013)

Phil. Mag. 94, 32–33 (2014) 35 PRB 91, 205103 (2015)





Proposal: Order Parameter is half-integer.



P. Coleman, R. Flint and PC, Nature 493, 611 (2013)

Phil. Mag. 94, 32–33 (2014) PRB 91, 205103 (2015)



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Proposal: Order Parameter is half-integer.

$\psi = \sqrt{\text{Multipole}} = \text{Spinorial OP}$

(Fractionalization of the OP)



P. Coleman, R. Flint and PC, Nature 493, 611 (2013)

Phil. Mag. 94, 32-33 (2014) 35 PRB 91, 205103 (2015)







P. Coleman, R. Flint and PC, Nature 493, 611 (2013) Phil. Mag. 94, 32–33 (2014) 35 PRB 91, 205103 (2015)



PRL 114, 236401 (2015)

week ending 12 JUNE 2015

Spectroscopic Determination of the Atomic *f*-Electron Symmetry Underlying Hidden Order in URu₂Si₂

L. Andrew Wray,^{1,2,3,*} Jonathan Denlinger,³ Shih-Wen Huang,³ Haowei He,¹ Nicholas P. Butch,^{4,5} M. Brian Maple,⁶ Zahid Hussain,³ and Yi-De Chuang³

The low-temperature hidden-order state of URu_2Si_2 has long been a subject of intense speculation, and is thought to represent an as-yet-undetermined many-body quantum state not realized by other known materials. Here, x-ray absorption spectroscopy and high-resolution resonant inelastic x-ray scattering are used to observe electronic excitation spectra of URu_2Si_2 , as a means to identify the degrees of freedom available to constitute the hidden-order wave function. Excitations are shown to have symmetries that derive from a correlated $5f^2$ atomic multiplet basis that is modified by itinerancy. The features, amplitude, and temperature dependence of linear dichroism are in agreement with ground states that closely resemble the doublet Γ_5 crystal field state of uranium.



Buhot et al., PRL (2014) Kung et al., Science (2015)

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Most significant T-dependence at the HO transition is in the A_{2g} channel



Constraints on theory?

Raman

Buhot et al., PRL (2014) Kung et al., Science (2015)



Scales nicely with c-axis magnetic susceptibility !!

J_z transforms under A_{2q} !!

Beautiful spectroscopy of $~\chi^{''}(q=0,\omega)$ consistent with previous neutron results at finite wavevector (analogous to SANS)

Buhot et al., PRL (2014) Kung et al., Science (2015)

Crystal-field Hamiltonian expanded to linear order in the electromagnetic stress tensor (in tetragonal environment)

$$H = \hat{H}_0 + \hat{O}_{A2g}(A_x A'_y - A_y A'_x)$$

where

$$\hat{O}_{A_{2g}} = \begin{bmatrix} a(\omega)(J_z^2 - J_y^2)J_xJ_y + b(\omega)J_z \end{bmatrix}$$

oscillatory field components of stress-energy tensor

Poynting vector

More work to be done to determine which term is larger (particularly in the presence of large spin-orbit coupling)

Nernst Effect

Colossal thermomagnetic response in the exotic superconductor URu₂Si₂

T. Yamashita¹, Y. Shimoyama¹, Y. Haga², T. D. Matsuda³, E. Yamamoto², Y. Onuki^{2,4}, H. Sumiyoshi¹, S. Fujimoto⁵, A. Levchenko⁶, T. Shibauchi^{1,7} and Y. Matsuda^{1*}



Figure 2 | Anomalously large Nernst signal and thermomagnetic figure of merit. a, The *T*-dependence of v (left scale) and ρ_{xx} (right scale) measured at $\mu_0 H = 1$ T near the superconducting transition. Both v and ρ_{xx} vanish at the vortex lattice melting transition temperature T_{melt} . **b**, Comparison of the v(T) data at $\mu_0 H = 1$ T between samples with different scattering rates (RRR = 1,080, 620 and 30). The data for RRR ~ 30 (expanded in the inset) is taken from ref. 23. **c**, Thermomagnetic figure of merit $ZT_e = N^2 \sigma T/\kappa$ at 1.5 K as a function of field in crystal #1 of URu₂Si₂ (red diamonds), compared with previous data in the semimetals PrFe₄P₁₂ (blue line) and Bi (black line) at 1.2 K taken from ref. 24.



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Is this giant Nernst effect a signature of exotic superconductivity or is it telling us something crucial about the Hidden Order phase?



Conclusions (for now)

• Recent Raman, elastoresistivity and RIXS measurements emphasize the importance of the Ising response, the multicomponent nature of Hidden Order parameter and the Gamma_5 U ground-state.....this all combined with the previous observation of Ising quasiparticles continues to suggest a spinor order parameter. Hidden order parameter has Ising anisotropy.

• Previous microscopics (Hastatic 1.0) must be revised to improve band structure (details of conduction electrons) and modelling of AFM phase (f-f hopping)

• Experiments we'd love to see:

Knight shift as a function of angle dHvA on all the heavy fermi-surface pockets Spin zeroes in the AFM phase (finite pressure) Low temperature probes of the 5f valence

> P. Coleman, R. Flint and PC, Nature 493, 611 (2013) Phil. Mag. 94, 32–33 (2014) PRB 91, 205103 (2015)

Broader Implications of Hastatic Order : A New Kind of Landau Order Parameter ?

Conventionally Landau theory in electronic systems based on formation and condensation of two-body bound states



e.g. s-wave superconductivity

When two-body bound-state wavefunction carries quantum number (spin, charge..), symmetry broken

All order parameters are bosons with integer spin !!

A new column in the classification of order parameters ??

Hastatic order generalizes Landau's concept to three-body bound-states

(Natural in heavy fermions, for the conventional Kondo effect is the formation of a three-body bound-state between a spin flip and conduction electron. However, in the conventional Kondo effect, the three body wavefunction carries no quantum number and is not an order parameter.)



Bound state of three fermions where the resulting fermionic bound-state carries integer spin while its 3-body wavefunction has 1/2 integer spin

(non-relativistic)

Hastatic order transforms under a double-group representation of the underlying symmetry group......order parameter fractionalization !!

Open Questions

• For Hastatic Order of URS

Development of Hastatic 2.0....new predictions ??

- Emergence of Superconductivity from the HO State?
- Other examples of Hastatic Order? (non-Kramers doublets in Pr and U materials)
- Direct Experimental Probe of Double-Time Reversal?
- Broader Implications: a new kind of broken symmetry where the order parameter transforms under a DOUBLE GROUP (S = 1/2) representation (symmetry analysis and Landau theory ??)





Thank you !!!