Exotic Structure, Exotic Ground States?

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Outline

• Frustrated magnetism in quasicrystals

- The "basics"
- Common features of magnetic quasicrystals
- Spin-glass-like behavior in magnetic quasicrystals

Superconductivity

- The "basics"
- Only a few claims of SC in quasicrystals.
- SC in i-Mg-Zn-Al and friends
- Why aren't there more examples?
- Unconventional SC
 - Heavy Fermion systems
 - Quantum Critical behavior in quasicrystals





Once the first two spins align antiparallel, the third one is *frustrated* because its two possible orientations, up and down, yield the same energy. Also true for polygons with odd number of edges (e.g. pentagons)

There are six nearest-neighbor interactions, four of which are antiparallel, but two of which are unfavorable.

2D: kagome lattice

3D: pyrochlore lattice

 $\mathcal{H} = J \sum \mathbf{S}_i \cdot \mathbf{S}_j$ pairs





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Magnetic Frustration (competing interactions)

$$H = -\sum_{i,j} J_{ij} \,\overrightarrow{S_i} \cdot \overrightarrow{S_j}$$

For AF interactions, J_{ij} is negative; For FM interactions, J_{ij} is positive. For particular values of, say, J_2/J_1 , we get frustration.

How do you get this?

- Distance dependent magnetic interactions such as the RKKY interaction.
 - $J_1 J_2$ or $J_1 J_2 J_3$ models where the different J_n can be FM or AF.
 - Disorder or dilution of the magnetic lattice.
- Different local environments of the magnetic ions lead to different interactions between them.
 - In the Al-based quasicrystals, for example, Only a small fraction of the TM carry a moment; distributed randomly on the quasilattice.



What kind of ground states can you get?

- Lattice distortion: Favors a particular magnetic ground state.
- Spin glass: A collection of spins whose low temperature state is a frozen and disordered. They feature a large number of degenerate ground states for the magnetic configurations.
- *Spin liquid*: A collection of spins whose low temperature state is disordered and exhibits strong quantum fluctuations even at 0 K.
- Spin ice: Large number of low energy states and residual entropy (like water ice).
 - Generally Ising-like single-ion anisotropy and FM interaction between spins on a tetrahedral cluster.
 - Long-range dipole interactions are important.
 - Novel spin-excitation spectra
- Exotic non-collinear spin arrangements/novel ground states
 - e.g nematic ordering in the iron pnictide superconductors



Antiferromagnetic order in quasicrystals?

- A great deal of theoretical work:
 - Magnetic interactions on a quasilattice. Non-trivial magnetic ordering is possible.
 - C. Godrèche, J. M. Luck and H. Orland, J. Stat. Phys. 45, 777 (1986).
 - R. Lifshitz, PRL 80, 2717 (1998); Rev. Mod. Phys. 69 1181 (1997).
 - S. Wessel, A. Jagannathan, and S. Haas, PRL 90, 177205 (2003).
 - E. Y. Vedmedenko, U. Grimm, and R. Wiesendanger, PRL 93, 076407 (2004).
- AI -TM (TM = 3d transition metal, e.g. Mn)
 - Only a small fraction of the TM carry a moment; distributed randomly on the quasilattice. Expect and see spin-glass-like behavior.
 - R. C. O'Handley, R. A. Dunalp and M. E. McHenry, Phil. Mag. B **61** 677 (1990).
 - F. Hippert and J. J. Préjean, Phil. Mag. 88, 2175 (2008).
- Rare-earth (local moment) alloys (e.g. *i-R*-Mg-Zn and *i-R*-Cd)
 - T. J. Sato, Acta Cryst. A 61, 39 (2005).
 - J. Dolinšek and Z. Jagličić, JAST **3**, 10 (2012).
 - A. I. Goldman, Sci. Technol. Adv. Mater. 15, 044801 (2014).







i-R-Mg-Zn



- Face-centered icosahedral quasilattice
- Bergman-type atomic clusters
 - R and Mg occupy the vertices of pentagonal dodecahedra forming an edge-sharing linked network
- Rhombohedral, hexagonal and cubic approximants of i-Y-Mg-Zn have been found but in most instances, magnetic properties of magnetic *R*-containing systems have not been fully studied.



Why study *i-R-Cd*

- Binary rather than ternary system
 - Absence of chemical "disorder"?
- Full series of heavy R systems
 - R = Gd, Tb, Dy, Ho, Er, Tm and Y
 - Systematic studies across the series of structure and magnetism
- AFM ordering in the 1/1 cubic approximant RCd_6 .
 - Differences in magnetic properties of periodic and aperiodic systems.
 - Manifests AFM long-range order.



RCd₆ structure





- Above T_S structure can be described as a body-centered cubic packing of Tsai clusters. (*a* ~ 15.5 Å)
- Transformation to monoclinic structure below $T_{\rm S} \sim 150 200$ K.
- Transformation triggered by orientational ordering of the inner tetrahedra within the cluster.

R. Tamura, Isr. J. Chem. 51, 1263 (2011)



RCd₆ ... AFM in a quasicrystal approximant

System	$\mu_{ m eff}$	$\mu_{ m calc}$	Θ	$T_{\rm N1}$	$T_{\rm N2}$	$T_{\rm N3}$	$T_{\rm N4}$	Reference
	$\left(\mu_{\rm B}/R \text{ ion}\right)$	$\left(\mu_{\rm B}/R \text{ ion}\right)$	(K)	(K)	(K)	(K)	(K)	
PrCd ₆	3.65	3.58	-11.33	0.13		_	_	[49]
NdCd ₆	3.55	3.62	-5.75	4.8	2.5			[49]
SmCd ₆	_	0.84	_	12.2	9.0	5.7	_	[49]
	0.59	0.84	-16	12.5	10.2	6.5	_	[47]
GdCd ₆	7.94	7.94	-32	18.9	13.2	7.3	2.5	[49]
TbCd ₆	9.3	9.72	-18	22.4	17.6		<u> </u>	[49]
	9.8	9.72	-17	24	19	2.4	_	[47]
DyCd ₆	10.9	10.63	-5.1	17.8	_	_	_	[49]
HoCd ₆	10.5	10.60	-1.0	8.4	6.8	3.4	_	[49]
ErCd ₆	9.1	9.59	-0.9	2.8	_	_	_	[49]
$TmCd_6$	7.4	7.57	-3.1	2.2	_	_	—	[49]



R. Tamura *et al.*, PRB **82**, 220201(R) (2010).
R. Tamura *et al.*, PRB **85**, 014203 (2012).
A. Mori *et al.*, *J. Phys. Soc. Jpn.* **81**, 024720 (2012).
M. G. Kim *et al.*, PRB **85**, 134442 (2012)
A. Kreyssig *et al.*, Phil. Mag. Lett. **93**, 512 (2013).



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TbCd₆ – Neutron diffraction on single crystals



- > Additional magnetic Bragg peaks at positions with H and K = integer + $\frac{1}{2}$
- Monoclinic structure with doubling of unit cell -> AFM order follows orientational order of inner Cd-tetrahedra
- XRMS measurements (not shown) show that for these half-integer magnetic peaks, the moment is along the monoclinic b-axis.







Spin-glass-like freezing rather than long-range magnetic order.

J. Dolinšek and Z. Jagličić, JAST **3**, 10 (2012). T. J. Sato *et al.*, J. Phys. Condens. Matter. **13** L105 (2001) I. R. Fisher *et al.*, J. Alloys Cmpd. **33**, 223 (2000). T. Kong *et al.*, Phys. Rev. B **90**, 014424 (2014).



Quasicrystals -- Quite similar magnetic properties

System	$\mu_{ ext{eff}}$	$(\mu_{\rm B}/R \text{ ion})$	$\mu_{\rm calc} \ (\mu_{\rm B}/R)$	ion)	Θ (K)	$T_{\rm f}$ (K)	$T_{\rm irr}$ (K)	Reference
Gd-Mg-Cd		7.9	7.94		-37	4.8	13	[1]
Gd-Mg-Cd		7.24			-37.8	4.3	_	[2]
Gd-Cd		—			-41	4.6	_	[3]
$\mathrm{Tb_8Mg_{42}Zn_{50}}$		10.05	9.72		-26	5.8	_	[4]
$\mathrm{Tb_9Mg_{34}Zn_{57}}$		9.91			-26.3	5.8		[5]
Tb-Mg-Cd		10.03			-23	5.6	12.5	[1]
Tb-Mg-Cd		9.74			-24.5	5.9	_	[2]
Tb-Cd		—			-21	5.3	8.7	[3]
$\mathrm{Dy_8Mg_{42}Zn_{50}}$		9.78	10.63		-17.2	3.8		[4]
Dy-Mg-Zn		10.5			-14.8	3.6	_	[5]
Dy-Mg-Cd		10.67			-14	3.8	7.4	[1]
Dy-Mg-Cd		10.59			-18.4	3.2		[2]
Dy-Cd		—			-11	3.0	10.1	[3]
$\mathrm{Ho_8Mg_{42}Zn_{50}}$		9.79	10.60		-10	<2	—	[4]
Ho-Mg-Zn		10.4			-7.8	1.95		[5]
Ho-Mg-Cd		10.42			-7	5.0	12.5	[1]
Ho-Cd		—			-6	1.76	—	[3]
$\mathrm{Er}_{8}\mathrm{Mg}_{42}\mathrm{Zn}_{50}$		9.59	9.59		-6.3	<1.5	—	[4]
Er-Mg-Zn		9.49			-5.1	1.30	—	[5]
Er-Mg-Cd		9.71			-6	4.4	—	[1]
Er-Cd		—			-4	1.11	_	[3]
Tm-Mg-Cd		7.08	7.57		-2	-		[1]
Tm-Cd		—	7.57		-2	0.63	_	[3]

Even though the structures (primitive vs. FCI, binary vs. ternary) and clusters (Bergman vs. Tsai) are different.

[1] Sato T J, Guo J Q and Tsai A P 2001 J. Phys. Condens. Matter **13** L105.

[2] Sebastian S E, Huie T, Fisher I R, Dennis K W and Kramer M J 2004 Philos. Mag. **84** 1029.

[3] Kong T, Bud'ko S L, Jesche A, McArthur J, Kreyssig A, Goldman A I and Caneld P C, Phys. Rev. B **90**, 014424 (2014).

[4] Charrier B and Schmitt D 1997 J. Magn. Magn. Mater. **171** 106.

[5] Fisher I R, Cheon K O, Panchula A F, Caneld P C, Chernikov M, Ott H R and Dennis K 1999 Phys. Rev. B **59** 308.



What's Freezing?

T. J. Sato, Acta Cryst. A 61, 39 (2005).



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i-Tb-Cd magnetic diffuse scattering

The scale of the features suggests that the magnetic correlations on the length scale of a single Tb-icosahedron are most important.





Spin-glass-like magnetic behavior rather than AFM order

- Why?
 - Structure?
 - Do the icosahedral clusters of *R* ions remain intact in the icosahedral phase?
 - Sources of disorder?
 - Magnetic interactions?
 - Local frustration introduced by aperiodicity?
 - Range of the RKKY interaction?
 - Something else?





- There are sizeable differences in composition between the *i*-*R*-Cd quasicrystals (e.g. GdCd_{7.9}) on one hand and the *i*-YbCd_{5.7} and *R*Cd₆ approximants on the other.
- Note: This is also true for *i*-ScZn_{7.33}

Question(s):

- Do the icosahedral *R* clusters remain intact in the quasicrystal?
- What is the origin of the lower *R* content?



The R site occupancy is not preserved



- (1) Derived compositions are reasonably close to the measured compositions
- (2) One of the sites in the Friauf polyhedra is occupied by *R*, whereas the other is fully or partially occupied by Cd, depending on the specific *R*.
- (3) The icosahedron is *not* fully occupied by *R* ions (only about 80%).

T. Yamada et al., *PRB* **94**, 060103(R) (2016).



How does this affect magnetic ordering?

Does this "disorder" explain the spinglass-like behavior?





Magnetic order persists.....but dominated by incommensurate magnetic order...**The search goes on.**



Let's try a simple model for moments on a cluster

- Try to model the spin arrangements on clusters with the following assumptions
 - Moments are Ising-like directed along the local 5-fold axes of the cluster (so there are 2¹² spin configurations).
 - All Tb moments (at low temperature) have their full value.
- What are the lowest energy spin configurations on a cluster for:

$$H=-\sum J_{ij}\,\vec{J}_i\cdot\vec{J}_j$$





Optimization yields:





Comparison of the simulations with magnetic diffuse scattering





Summary (Magnetic frustration)

- We now have a number of examples of binary and ternary magnetic quasicrystals and a corresponding 1/1 approximants to work on.
- Magnetic frustration undoubtedly plays a role in these systems.
- But the origin of spin-glass-like behavior in quasicrystals remains an open question.
 - Structure?
 - Quasicrystallinity and/or "disorder"
 - Magnetic interactions?
 - Local frustration?
 - Range of the RKKY interaction?
 - Something else?

• But we are starting to close in on the answers!

See talks by: A. Koga, T. J. Sato and R. Tamura in Session 2 on Monday, and T. J. Sato in Session 7 on Wednesday.



Superconductivity (Zero Resistivity)

- Superconductivity refers to a material state in which current can flow with **no** resistance below the superconducting transition temperature, T_c.
- Resistance in a conductor stems from scattering of electrons from thermally activated ions.
 - Resistance generally decreases as temperature decreases.
 - The decrease in resistance in normal metals reaches a minimum based on disorder and impurities in the lattice.



Superconductivity in mercury (1911).



Superconductivity (Meissner Effect)

Type I superconductors are characterized by the Meissner effect, i.e. flux is fully expulsed through the existence of supercurrents

Type II superconductors find it energetically favorable to allow flux to enter via normal zones termed vortices.





- H_c: Thermodynamic critical field
- $\rm H_{c1}:$ critical field defining the transition from the Meissner state

 H_{c2} : Critical field defining the transition to the normal state

U. Essmann and H. Trauble, Physics Letters 24A, 526 (1967).



Conventional superconductivity

BCS theory starts from the assumption that there is some attraction between electrons, which can overcome their Coulomb repulsion.

An electron moving through the lattice "deforms" the lattice causing another electron, with opposite spin, to move into the region of higher positive charge density \rightarrow correlated electrons. This is the essence of the electron-phonon interaction.

Because there are many such electron pairs in a superconductor, and they are correlated because of the Pauli exclusion principle, breaking one pair requires changing the energy of all other pairs.

 An "s-wave gap" is formed at E_F which vanishes like a second order phase transition at T_C:

 $\Delta(T = 0) = 1.764 k_{\rm B} T_{\rm C}$

• $T_{\rm C}$ can be cast in terms of the electron-phonon coupling strength, *V*, and the Debye frequency, $\omega_{\rm D}$, and the electronic density of states at $E_{\rm F}$, $N(E_{\rm F})$:

 $k_{\rm B}T_{\rm C} \sim \hbar\omega_{\rm D} {\rm e}^{-1/N(E_{\rm F})V}$

• The specific heat shows a "jump" at T_c and an exponential decrease below with the opening of the superconducting gap.



Superconductivity in a quasicrystal approximant





A relevant example: Au-Ge-Yb 1/1 approximant K. Deguchi *et al.*, J. Phys. Soc. Jpn. **84**, 023705 (2015)

- ρ = 0 below $T_{\rm C}$ = 0.68 K and 0.36 K for AGY(I) and AGY(II), respectively.
- Full Meissner effect \rightarrow bulk superconductivity
- Specific heat jump at T_C and exponential decrease below.

 $\begin{array}{l} \mathsf{AGY(I):} \ \mathsf{Au}_{64.0}\mathsf{Ge}_{22.0}\mathsf{Yb}_{14.0} \\ \mathsf{AGY(II):} \ \mathsf{Au}_{63.5}\mathsf{Ge}_{20.5}\mathsf{Yb}_{16.0} \end{array}$

Both samples are composed of Tsai-type clusters in a bcc packing, but AGY(II) features a single Yb ion at the center of the cluster rather than a tetrahedron of Au, which may carry a magnetic moment.

There are not many reports of SC in Qxtals.

- SC claimed in both the *i*-phase (0.81 K) And FK phase (0.73 K) of Mg-Cu-AI, but no magnetization data presented.
 K. M. Wong *et al.*, Phys. Rev. B 35, 2494 (1987).
- And the *i*-phase (0.72 K)and FK phase (0.81 K) of Al-Li-Cu, but again no magnetization data and some contamination of *i*-phase

➢ J. L. Wagner *et al.*, Phys. Rev. B **38**, 7436 (1988).

And the *i*-phase of Ti-Zr-Ni (<1.4 K), but again no magnetization measurements and multiphase samples.

➢ V. Azhazha *et al.*, Phys. Lett. **A303**, 87 (2002).

And the *i*-phase of Al₆Ru, but again no magnetization measurements.

S. N. de Medeiros *et al.*, Ferroelectrics **305**, 193 (2004).

The major worry here is about filamentary SC from related phases!



SC in quasicrystalline Mg-Zn-Al?

E. Graebner and H. S. Chen, PRL 58, 1945 (1987).

TABLE I. Numerical results for resistivity ρ , specific-heat fitting parameters γ , β , α , η , T_c , and ΔT_c in Eqs. (1) and (2), the electronic specific-heat ratio C_{es}/C_{en} at T_c , and derived quantities Θ_D (Debye temperature), v (sound velocity), λ (electron-phonon coupling parameter), and N(0) (unrenormalized density of electronic state). N(0) is in units of states/eV atom.

	Cubic	Icosahedral	Amorphous
$\rho_{300 \text{ K}}$ ($\mu \Omega \text{ cm}$)	90	79	380
P4.2 K	71	77	400
T_c (resistivity) (K)	0.330	0.442	1.25
ΔT_c (resistivity)	0.015	0.020	0.15
$\gamma (10^{-5} \text{ J/g K}^2)$	2.3 ± 0.2	2.2 ± 0.3	1.9 ± 0.3
$\beta (10^{-6} \text{ J/g K}^4)$	1.12 ± 0.04	2.75 ± 0.3	7.5 ± 0.6
$\alpha (10^{-5} \text{ J/g K}^2)$	0.003	0.06	0.3
$\eta (10^{-5} \text{ J/g K})$	0.86 ± 0.15	1.20 ± 0.2	1.8 ± 0.4
T_c (specific heat) (K)	0.315 ± 0.01	0.410 ± 0.01	0.754 ± 0.03
ΔT_c (specific heat) (K)	0.026	0.050	0.18
$C_{es}/C_{en}(T_c)$	2.9 ± 0.6	3.0 ± 0.8	3.1 ± 0.8
$\Theta_{\rm D}({\rm K})$	348	258	185
v (10 ⁵ cm/sec)	3.10	2.28	1.6
λ	0.36	0.39	0.45
N(0)	0.15	0.14	0.11



TEMPERATURE (K)

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But.....

No SC observed in I or Frank-Kasper (FK) phases of Mg_{39.5}Zn_{40.3}Al_{20.2} down to 0.3 K.
 K. M. Wong *et al.*, Phys. Rev. B **35**, 2494 (1987).

- SC not found in the I-phase of Mg-Zn-AI
 K. Kamiya *et al.*, Abstract for the Toyota RIKEN International Workshop (2015).
- Possibility of second phase contamination by 1/1 approximant
 K. Kamiya *et al.*, *Nature Commun.* 9, 154 (2018)

So, what's going on here?



SC in Al-Zn-Mg quasicrystal and approximants



K. Kamiya *et al.*, *Nature Commun.* **9**, 154 (2018)



- $T_{\rm C}$ for the 1/1 approximants vary from about 0.8 K \rightarrow 0.2 K.
- T_C for the 2/1 approximant and quasicrystal are quite similar (~ 50 mK)



SC in Al-Zn-Mg quasicrystal and approximants







- It would be lovely, but tough, to get M vs. T at lower temperatures.
- Jump in C_{e} for both the 1/1 approximant and the quasicrystal are consistent with BCS.
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Why is $T_{\rm C}$ so low for the quasicrystal?

• Within the context of BCS theory we have some choices:

 $k_{\rm B}T_{\rm C} \sim \hbar\omega_{\rm D} {\rm e}^{-1/N(E_{\rm F})V}$





• Recall that qxtals are characterized by a pseudogap in the DOS at the Fermi energy.



- Why aren't there more reports of SC in quasicrystals?
 - Practical limitation?: How low in temperature can your PPMS or MPMS system go?
- Where to look?
 - Matthias' rules (pre-high $T_{\rm c}$)
 - high symmetry is good, cubic symmetry is the best
 - high density of electronic states is good
 - stay away from oxygen
 - stay away from magnetism (e.g. 3d transition metals)
 - stay away from insulators
 - stay away from theorists!!



Summary (Superconductivity)

- Reinvestigation of previous observations of SC
 - Do we see full diamagnetic response and specific heat anomaly at $\rm T_c?$
- Take a second look at some of our old friends (Al-based QCs)
 - Collect enough cases to establish common threads among different "classes" of quasicrystals in comparison to crystalline and amorphous phases.
- The observation of SC in the Au-Ge-Yb 1/1 approximant and the Al-Zn-Mg quasicrystal should stimulate further investigations.
- What about unconventional superconductivity?

See talks by: A. Jagannathan, N. Takemori, S. Sakai in Session 4 on Monday.



Unconventional superconductivity

- Symptons:
 - For BCS theory, since the electron-phonon interaction is key, the absence of an isotope effect, where $T_{\rm C}$ varies with the mass of the isotope, is one indicator that something funny is going on.
 - $T_{\rm C}$ predicted by BCS theory is too low.
 - Unconventional SC often occurs close to the boundary with magnetic order.
 - Generally held that the SC arises from strong magnetic fluctuations, often associated with a QCP.





Heavy Fermions

- Generally contain rare earth or actinides which form a lattice of localized magnetic moments.
- In the single ion Kondo effect: Local moments develop an AFM coupling with the spins of the conduction electrons:

$H_I = J \sigma \cdot S$

effectively screening the local moments below the Kondo temperature, T_{k} .

- In a "Kondo Lattice", there is competition between the RKKY interaction promoting AFM order and the Kondo screening.
- Below a characteristic temperature, T^{*}, a Fermi Liquid of "heavy electrons" arises from the local moment-conduction electron interaction with effective masses 100-1000 times the free electron mass.



Heavy Fermions and Quantum Criticality



One can drive the system via pressure, magnetic field or doping to a point where the AFM ordering is driven continuously to zero temperature.

A quantum critical point (QCP) separates the Heavy Fermion and AFM ordered regimes.

"Doniach Diagram"



Quantum Critical State in Au-Al-Yb

- Generally one has to "tune" a system to a QCP via pressure, doping or magnetic field
 - i-Au-Al-Yb manifests NFL behavior at zero field and ambient pressure (i.e. $\rho(T) \sim T$, C/T \sim -ln(T)).
 - The approximant phase exhibits Heavy Fermion behavior at low temperature (i.e. $\rho(T) \sim T^2$, C/T saturates to 0.7 J/K-mol-Yb).
 - The quantum critical state is robust with pressure.



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K. Deguchi *et al*., Nature Mat. **11**, 1013 (2012).

Looking Forward

- The observation of quantum critical behavior in Au-Al-Yb, and differences with the 1/1 approximant are intriguing
 - Other examples?
 - Look for HF and QC behavior in other Yb, Ce, U or Pu (any volunteers?) compounds.
 - The observation of quantum critical behavior in Au-Al-Yb, and differences with the 1/1 approximant are intriguing, particulary the robust behavior with pressure.
 - Search for unconventional SC in quasicrystals close to a QCP

See talks by: K. Imura, S. Watanabe and T. Ishimasa in Session 8 on Wednesday, and K. Deguchi in Session 10 on Thursday.



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Thank you for your attention.



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