Department of Physics
Condensed Matter Physics
Clarendon Laboratory, Parks Road, Oxford OX1 3PU

UNIVERSITY OF OXFORD

CONDENSED MATTER SEMINAR

Thursday 23rd of February at 2.15pm

"Neutron Spectroscopy on the Most Complex Element: Plutonium"

Dr Marc Janoschek
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Magnetism is typically described in one of two extreme limits: In insulators electrons are treated as localized objects which carry a magnetic moment, whereas in the metallic regime magnetism is generally considered to arise from an instability of the Fermi surface. Strongly correlated electron systems typically display a duality between these two pictures. Electrons behave both itinerantly giving metallic like behavior, yet can also possess a large fraction of their spectral weight in high energy Hubbard-like bands that are more local in nature.
This duality has been intensively studied from the charge perspective and continues to be an active area of research. However, this duality also is manifest in the spin degrees of freedom, and therefore neutron spectroscopy represents a powerful tool to gain insight in such electronic dichotomy as demonstrated by our recent measurements on plutonium (Pu) metal. Pu is arguably the most complex elemental metal known because its 5f electrons are tenuously poised at the edge between localized and itinerant configurations. This complex electronic structure leads to emergent behavior—all a direct consequence of its 5f electrons— including six allotropic phases, large volumetric changes associated with these transitions of up to 25%, and mechanical properties ranging from brittle α-Pu to ductile δ-Pu. Pu also exhibits a Pauli-like magnetic susceptibility, electrical resistivity and a Sommerfeld coefficient of the specific heat that are an order of magnitude larger than in any other elemental metal. Finally, while experiments find no sign for static magnetism in Pu, most theories that use the correct volume predict a magnetically ordered state. We have investigated the magnetic fluctuation spectrum of δ-Pu using the ARCS spectrometer at SNS [1]. Our study reveals that the ground state of plutonium is governed by valence fluctuations, that is, a quantum-mechanical superposition of localized and itinerant electronic configurations. Our results that are in quantitative agreement with dynamical mean field theory show that the magnetism in Pu is not "missing" but dynamic, and is driven by virtual valence fluctuations. Our measurements provide a straightforward interpretation of the microscopic origin of the large, Pauli-like magnetic susceptibility of δ-Pu. Finally, because the various valence configurations imply distinct sizes of the Pu ion, the valence-fluctuating ground state also gives a natural explanation for its complex structural properties and notably the large sensitivity of its volume to small changes in temperature, pressure or doping.

[1] M. Janoschek, et al. "The valence fluctuating ground state of plutonium" Sci. Adv. 1, e1500188 (2015)

Host: Prof Andrew Boothroyd

Audrey Wood Seminar Room, Clarendon Laboratory