Experimental report

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Proposal: 5-31-2438 **Council:** 4/2015

Title: Quantum kagome lattice in a van der Waals solid

Research area: Physics

This proposal is a new proposal

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Samples: O2/N2

Instrument	Requested days	Allocated days	From	To
D20	4	4	26/11/2015	30/11/2015

Abstract:

We propose to measure a novel spin-1 kagome system in a van der Waals solid. The O2/N2 gas mixture with 48% oxygen solidifies into a hexagonal structure above 8 GPa pressure, where the O2 molecules build up stacked kagome lattice. This structure combined with the spin of the oxygen atoms and the expected antiferromagnetic exchange interaction between nearest neighbors makes this system an exciting model to study kagome physics. We propose to use neutron diffraction to determine the crystal structure down to 4 K and to measure the magnetic correlations.

Quantum kagome lattice in a van der Waals solid

We have recently carried out a very successful neutron diffraction study on a 50%-50% mixture of O_2 and N_2 ('solid air') on D20 which confirmed the existence of an O_2 kagome phase that was indirectly concluded from an X-ray diffraction study. The kagome phase has space group *P6/mmm* with the O_2 and N_2 molecules on different crystallographic sites. For the 50%-50% O_2-N_2 mixture, the magnetic O_2 molecules (S=1) form a kagome lattice while the N_2 molecules are separating these layers. Since oxygen and nitrogen have very different neutron scattering length (unlike X-rays), we could determine the occupancy of the crystallographic sites. Our data show that the kagome plane is mostly occupied by oxygen. However due to the strong preferred orientation, we were not able to carry out a more detailed analysis such as determining the anisotropic thermal displacements. The axis of each O_2 molecule point along the *c*-direction, thus we expect frustrated antiferromagnetic exchange within the kagome layers.

We also mapped out the pressure-temperature phase diagram, which revealed the existence of the hexagonal phase below 2 GPa at low temperatures, and discovered a new low temperature monoclinic phase. This phase is stable below 89 K at 4.9 GPa and probably also stable at ambient pressures if the temperature is low enough. In the diffraction pattern of this new phase only reflections with both non-zero inplane and out-of-plane components are split. By fitting the split peak positions, we could determine the new monoclinic cell parameters, for example at $T = 6$ K, $p = 7.5$ GPa: a = 5.679(11) Å, b = 9.716(47) Å, c = 8.276(27) Å, $\beta = 49.93(12)$ °. From the $b/a = 0.988\sqrt{3}$ ratio we conclude that the *ab*-plane is very weakly perturbed by the hexagonal to monoclinic phase transition, only the layers are shifted relative to each other. This means that the frustrated 2D kagome geometry of the O_2 molecules is kept at low temperatures. This result is important in light of the behavior of pure O_2 where the magnetically frustrated rhombohedral β-phase distorts at low temperature to release magnetic frustration. We did so far not find any sign of long-range magnetic order in the monoclinic phase down to 6 K. This can be the sign of frustration and low dimensionality.

To reach the necessary pressures we used the ILL high-pressure gas loader to load the samples at ambient temperature and 2-3 kbar. This leads to sufficient starting density in order to reach 10 GPa under further compression in the Paris-Edinburgh cell on D20.