Experimental report

05/05/2016

Proposal: 7-05-449 **Council:** 4/2015

Title: Ferroelectric ordering in the endofullerene H2O@C60 and the effects of applied electric field

Research area: Physics

This proposal is a new proposal

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Samples: H2O@C60

Instrument	Requested days	Allocated days	From	To
IN5	6	6	29/10/2015	05/11/2015

Abstract:

This proposal concerns the translation-rotation quantum dynamics of water molecules encapsulated inside a C60 cage in the endofullerene H2O@C60 and the influence of the electric dipole moments on intra- and inter-cage interactions. For classical dipoles a transition to ferro- (or antiferro-) electric ordering is expected at temperatures below the interaction energy ~3K. In H2O@C60 the transition temperature will be lower as quantum rotational delocalization lowers the interaction energy. Studying the INS spectrum (in NE gain and loss) of a 100% filled sample at low temperature using the mK fridge, we shall investigate shifts in the energy of the ortho-H2O states that will accompany the ferroelectric ordering. We propose further investigating intra- and inter-cage interactions by studying changes that arise in the NE gain spectrum when an electric field is applied to partially align the electric dipoles.

Experiment n°: 07-05-449
Title: Ferroelectric ordering in the endofullerene $H_2O@C_{60}$ and the effects of applied electric field
Instrument: IN5
Dates of experiments: 29/10/2015 to 04/11/2015
Experimental Team: Tony Horsewill: School of Physics & Astronomy, University of Nottingham; Salvatore Mamone: School of Physics & Astronomy, University of Nottingham
Local Contacts: Stéphane Rols, J. Ollivier, Mark R. Johnson

Introduction

This proposal concerns the study of the effects of electric dipole moments on intra- and inter-cage interactions in water molecules confine inside C_{60} fullerenes at low temperatures with and without applied external electric field. When confined inside C_{60}, water molecules are isolated from each other allowing for unhindered quantum rotations at cryogenic temperatures.[1] Interestingly $H_2O@C_{60}$ is a real world example of dipolar quantum rotors in which electric dipoles may align cooperatively leading to the observation of a new class of ferroelectric materials.[2] INS is uniquely equipped to investigate properties of light molecules confined in fullerenes. The main features of the quantum dynamics of $H_2O@C_{60}$ have been elucidated in previous works.[1,3] This proposal is moving on the same ground with other proposals (,07-05-447, 07-04-149, 5-12-306) investigating the quantum motion, nuclear spin conversion phenomena and symmetry breaking effects in water@C_{60}.

Samples

$H_2O@C_{60}$ was synthesized in the laboratory via molecular surgery procedures.[4,5] For this IN5 experiment we used two samples synthesized by our collaborators from Southampton (led by Prof. R. J. Whitby)

- 88 mg with a filling factor of 99.6%;
- 100 mg with a filling factor of 10 %;

The filling factor refers to the percent of water-filled cages versus total cages in the sample and was determined by high performance liquid chromatography. The samples come from the same batch that was used for proposal 07-05-449 used as for proposal. Both samples were in powder form.

The 99.6% filled sample was used for experiments at mK temperatures using the $^3He/^4He$ dilution fridge available at ILL. The aim of the mK experiments was to observe variations in the INS line that could be related to electric ordering at low temperatures.

The 10% filled sample was used for experiments with applied electric field at temperatures down to the base temperature of the orange ILL cryostat (1.6K). The aim of the electric field experiments was to observe Stark shifts in order to provide a direct measurement of the electric dipole moment of the complex. For the experiments in the electric field the sample was placed between two aluminum plates separated by a 3.15mm thick Teflon spacer in which a rectangular hole of dimension 7mm by 5mm was created to accommodate the $H_20@C_{60}$ sample (~ estimated 100 mg).

Experiments

The temperature dependence of the INS transitions between the ortho ground state sub-levels and the non-degenerate para ground state of $H_2O@C_{60}$ is shown in figure 1. The energy level structure of $H_2O@C_{60}$ is discussed in the paper of Goh's et al. [1,3]**.** No observable change in the ortho-para ground state line (at ± 2.55 meV) is evident in the spectra.

The effect of applied electric field on the ortho to para ground state INS line is shown in figure 2. No Stark shift was observed up to the highest applied voltage (~12 kV) above which irreversible electric breakdown was observed.

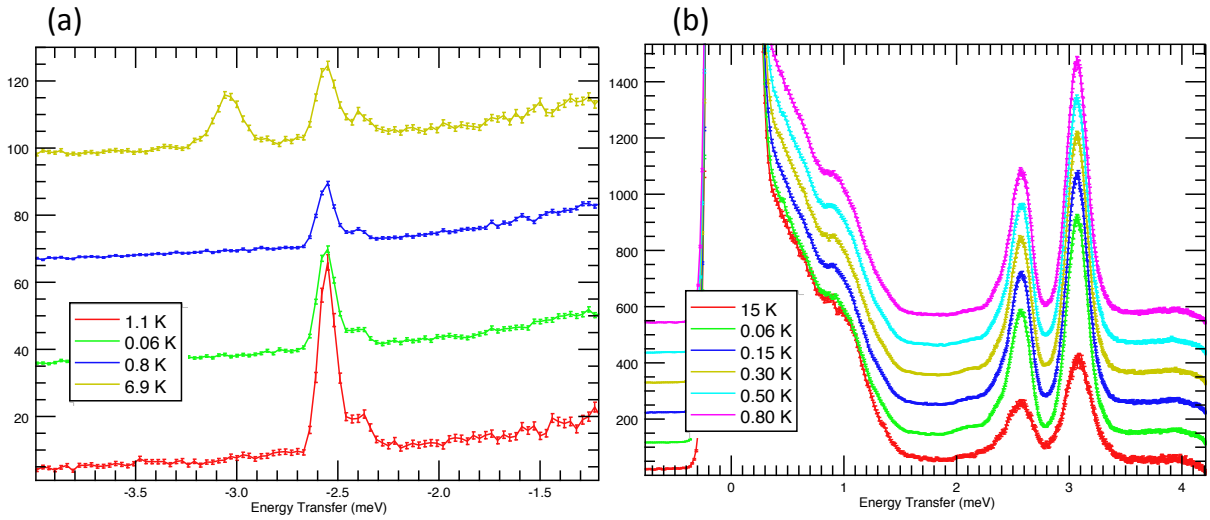

Figure 1 The temperature dependence of the INS spectra of $H_2O@C_{60}$ (99.6% filled) as collected on IN5 is shown (a) at 8Å in neutron energy gain and (b) at 3.6Å in neutron energy loss. In panel (a) the spectra were acquired in the following chronological order: 1.1 K , 0.8 K, 0.06 K and 6.9 K. In panel (b) the spectrum at 15 K was acquired first and then all the spectra from 0.06K to 0.8 K were sequentially collected.

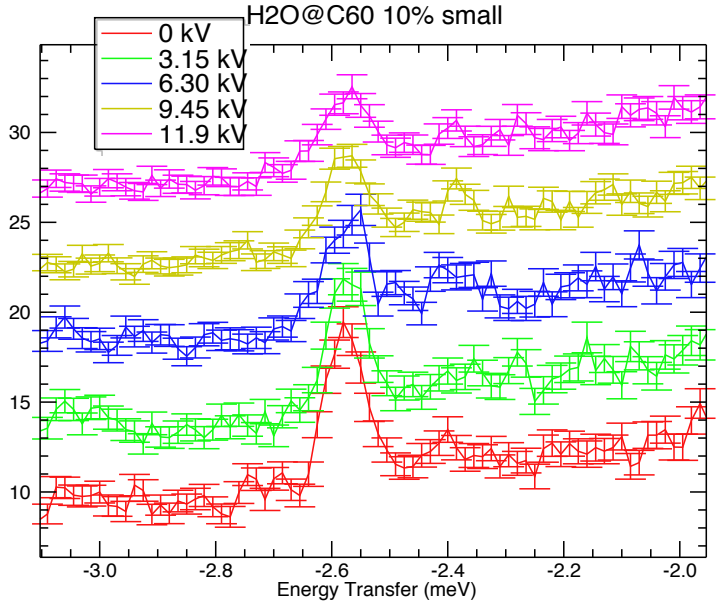

Figure 2 Effect of applied electric field on the INS spectra of $H_2O@C_{60}$ (10% filled) at 1.6 K as collected on IN5 in neutron energy gain at 8 Å.

Bibliography

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