Experimental report

14/02/2017

Proposal: **4-01-1530** **Council:** 4/2016

Title: Evolution of Longitudinal Spin Fluctuations in Ca(2-x)SrxRuO4

Research area: Physics

This proposal is a new proposal

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Samples: Ca(2-x) Sr(x) Ru O(4) ; x = 0.2
Ca(2-x) Sr(x) Ru O(4) ; x = 0.1
Ca(2-x) Sr(x) Ru O(4) ; x = 0.15

Instrument	Requested days	Allocated days	From	To
IN8 Flatcone	10	5	07/12/2016	12/12/2016

Abstract:

Recently, research in solid state physics concentrates on the influence of spin-orbit coupling (SOC) in transition metal oxides (TMO). While systems with partially filled 5d-orbitals are determined by SOC, its effect on 4d elements is still under discussion. A novel theory [1] considers the magnetism in TMO systems with such intermediate strong SOC. Following its predictions, we performed inelastic neutron scattering experiments on the model system Ca2RuO4 and observed a magnetic dispersion that is distinct different to conventional magnetism. It features a longitudinal magnon mode, a defining aspect of the theory.
Having established this amplitude mode, we plan to follow its evolution upon Sr-substitution, and thus investigate the influence of SOC during the transition from an anti-ferromagnet to a metal. We propose to investigate the evolution of magnetic dispersion as a function of Sr-substitution, using the thermal triple axis spectrometer IN8 in its Flatcone modification.
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[1] G. Khaliullin PRL 111, 197201 (2013)

Experimental report

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Instrument: IN8, flatcone option

Introduction

For several decades ruthenium-based compounds have been a primary focus of condensed matter physics. They feature many phenomena such as the unconventional spin-triplet superconductivity of Sr_2RuO_4[1], the orbital ordering in $La_4Ru_2O_{10}$ [2] or the Haldane gap for the spin $S = 1$ system $Tl_2Ru_2O_7$ [3]. With many spin, orbital and lattice degrees of freedoms, these systems are determined by the intricate interplay of many different interactions. These entail spin-orbit coupling (SOC), magnetic exchange coupling, effects of the crystal field, the cooperative Jahn-Teller effect, and Hund's coupling.

A novel theoretical approach considers the van Vleck type Ru4+ ions in a crystal field [4]: Without SOC, low-spin t_{2g} electrons with nominally $S = 1$ moments interact via the Goodenough-Kanamori type superexchange interaction. However, if the SOC is much larger than the tetragonal crystal field splitting , the coupling to $L = 1$ orbital moments results in a singlet-triplet splitting with a $J = 0$ non-magnetic ground state (singlet) and $J = 1$ first excited state (triplet). In an intermediate regime, the compressive tetragonal distortion splits the triplet and lowers the cost to excite a triplon, making condensation into a magnetically ordered ground state possible. This results in a proposed unique spin excitation spectrum, and the unique material conditions in Ca_2RuO_4 enable the direct measurement of the defining (Higgs) amplitude mode [4].

Experiment 4-01-1530

setup IN8

Aim of the present experiment was to investigate the evolution of the magnetic excitations as the system is tuned towards the quantum critical point using inelastic neutron scattering at the triple axis spectrometer IN8. In this experiment we used the IN8 triple-axis spectrometer in its *flatcone* mode. The flatcone detector was configured to use the Si analyzer with $k_f = 3\,\text{Å}$, and by rotating the detector out of the horizontal plane, we were able to increase the reciprocal space coverage. We chose the new Si111 analyzer recently installed at IN8. Compared to the original setup, we profited from a higher overall neutron flux and reduced higher order contamination in the incident neutron beam. This setup enabled us to record very rapidly constant energy maps around the magnetic zone center.

Results

The $Ca_{2-x}Sr_xRuO_4$ system undergoes a first order structural transition for all $x < 0.2$. Here, the c-axis of the structure shrinks upon cooling by 3 %, leading to severe sample degradation during the transition from the so-called S- to the L-phase. While the transition temperature for the pristine compound is 357 K, it is rapidly suppressed by Sr substitution to temperatures lower than room temperature, making cooling of the samples very difficult. As a result, we took special care with respect to degradation issues of our sample, i.e. we enveloped the crystals with a hard shell of a hydrogen-free epoxy often used in the neutron scattering community for fixating crystals in a co-aligned array (CYTOP).

While we were able to preserve most crystals of our co-aligned array, the largest crystals degraded during cooling and the peak intensity was reduced by approximately 35 %. Nevertheless, measurements at different temperatures resulted in a reasonable contrast and we were able to follow the magnetic dispersion up to energies of 40 meV.

In our data-analysis we are considering absorption artifacts, and numerous spurions which are introduced by the fact that the flatcone detector can not be used with PG filters. We will furthermore fit the theoretical model develop by G. Khaliullin to the dispersion. Thus we estimate that the data will be fully analyzed by April 2017.

Outlook

We are investigating the implications of Sr substitution in the Ca_2RuO_4 system. Our first experiments on pristine calcium ruthenate resulted in the discovery of a longitudinal mode (Higgs mode) in a solid state system. Furthermore, we found the experiments with low Sr content to be challenging due to the degradation processes associated with a sub-room-temperature first oder phase transition. With this last experiment we are able to show that with Sr substitution the gap slightly closes, and the dispersion becomes much steeper.

For $0.2 < x < 0.5$, a magnetic metallic state that still features antiferromagnetic order[6] exists at low temperatures. At higher levels we expect to transition through the aforementioned quantum critical point into a paramagntic metallic regime. Our future experiments will investigate the magnetic dispersion for Sr substitution levels corresponding to the magnetic metallic state, in the vicinity of the aforementioned quantum critical point.

References

[1] Y. Maeno et al., Nature **372**, 6506 (1994)
[2] P. Khalifah et al., Science **297**, 5590 (2002)
[3] S. Lee et al., Nature **5**, 471 (2006)
[4] G. Khaliullin, PRL **111**, 197201 (2013)
[5] S. Nakatsuji, Y. Maeno, PRL **84**, 2666 (2000)
[6] S. Nakatsuji, Y. Maeno, PRB **62**, 6458 (2000)