Experimental report

25/01/2024

Proposal: **CRG-3001**

Council: 4/2023

Title: Anomalous spin dynamics in Ca-doped SrRuO3

Research area: Physics

This proposal is a new proposal

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Samples: $Ca_{0.3}Sr_{0.7}RuO_3$
$Ca_{0.5}Sr_{0.5}RuO_3$

Instrument	Requested days	Allocated days	From	To
IN12	6	6	25/10/2023	31/10/2023
ORIENTEXPRESS	1	1	05/11/2023	06/11/2023

Abstract:

Ferromagnetic $SrRuO_3$ exhibits an anomalous temperature dependence of the magnon gap and of the stiffness that could be attributed to the impact of Weyl points in the electronic structure. Here we propose to apply a similar analysis to two compounds with partial substitution of Sr by Ca. The Ca content reduces the ferromagnetic transition temperature as well as the ordered moment, while the size of the local magnetic anisotropy seems to little change. As Weyl points were already established also for these substituted materials we can deepen the analysis of the interplay of Weyl points and the topology of the electronic structure with the spin dynamics.

Experimental Report

Instrument	IN12
Proposal Number	CRG-3001
Proposal	Anomalous spin dynamics in Ca-doped $SrRuO_3$
Experimentalist	Zharasadat Ghazinezhad, Akshay Tewari, Markus Braden
Local Contact	Karin Schmalzl

$SrRuO_3$ (ordered moment of 1.6 μ_B, T_C = 165 K) has been established to be a ferromagnetic Weyl semi-metal, in which Weyl points not only imply peculiar magnetotransport properties [1,2] but also impact the spin dynamics [3,4]. The anomalous Hall effect in $SrRuO_3$ arises from the Berry phase around the Weyl points, and the renormalization of the magnon stiffness and of the magnon gap scales with the anomalous Hall effect [3,4]. Upon substituting Sr by Ca the ferromagnetic transition in $Sr_{1-x}Ca_xRuO_3$ gets suppressed and fully vanishes around x~0.7 [5]. The aim of this experiment was to study the magnetic excitations in two $Sr_{1-x}Ca_xRuO_3$ single-crystalline samples with x=0.3 and 0.5, that exhibit ferromagnetic transitions at 91 and 50 K, respectively. We used pyrolithic graphite crystals as monochromator and analyzer. To suppress higher order contaminations, we inserted a velocity selector and a Beryllium filter, and most scans were performed with fixed k_f=1.55Å^{-1}. All reciprocal space vectors are given in reduced lattice units with respect to the pseudocubic lattice of about 3.9Å.

Due to technical reasons, we started with the slightly larger higher-doped x=0.5 crystal but here we discuss the results in order of increasing Ca content. Fig. 1 shows constant energy scan at the ferromagnetic zone center (100). At low temperature there is evidence for a gap appearing at much higher energy (~2.6meV) than that in pure $SrRuO_3$ (at about ~1meV). This confirms the anomalous behavior in the pure material where Weyl points renormalize the gap [3]. Closer inspection of the data at 1.6 K indicates that the gap is further split into two components with an additional mode near 3.7 meV. A splitting of the anisotropy gap is expected for the orthorhombic system. Furthermore, both gaps shift to lower energies upon heating to the Curie temperature.

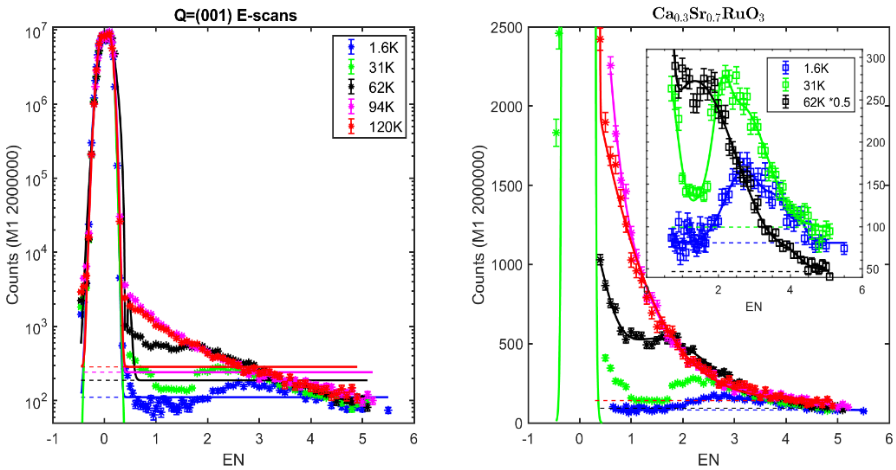

Fig. 1 Constant-***Q*** scans at the ferromagnetic zone center (100) for 30% of Ca doping. At low temperature one distinguishes two gap modes at rather high energies that decrease

When further analyzing constant-energy scans taken at 1.6 K one finds evidence for a finite signal below the gap, see Fig. 2. Furthermore, this agrees with the interpretation of the x=0.5 data, see below. For x=0.3 the ferromagnetic moment is already strongly suppressed to about ~0.5 μ_B, well below the value of the pure material, which can imply longitudinal scattering. This in-gap, possibly longitudinal scattering strongly increases with temperature, and near the Curie temperature the gaps are no longer visible in the quite strong paramagnon scattering. It is remarkable that the anisotropy gaps behave oppositely to the magnetization when comparing x=0 and 0.3, which underlines an exotic mechanism. Energy scans taken at 2.5 and 4 meV for different temperatures illustrate the picture that the entire magnon dispersion shifts to lower energies upon heating so that the paramagnon scattering already exhibits separated contributions at intermediate energies, whereas the scattering at 1.6 K is split.

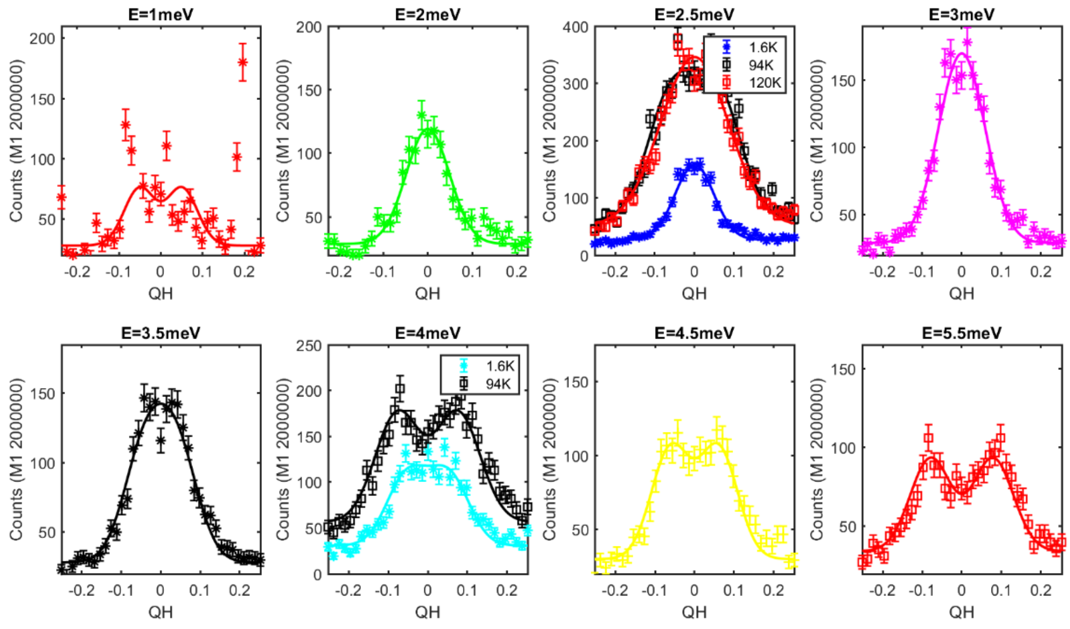

Fig. 2: Constant-energy scans across the magnon and paramagnon dispersion in $Sr_{0.7}Ca_{0.3}RuO_3$. While at low temperature the magnon dispersion only starts above the rather large gap values, separated modes already appear at much lower energy near and above the Curie temperature.

In the more strongly substituted $Sr_{0.5}Ca_{0.5}RuO_3$ we find a qualitatively equal dispersion, but the anisotropy gaps are further enhanced reaching 3 and ~4 meV at low temperature. The strange inverse relation with the ordered moment, which is further reduced at x=0.5 to ~0.4 μ_B, thus persists to this Ca concentration. Also, the temperature dependent filling of the energy range below the gap strongly resembles the findings for x=0.3. But for x=0.5 this process is enhanced due to the lower Curie temperature and due to the relatively stronger scattering by excitations below the gap.

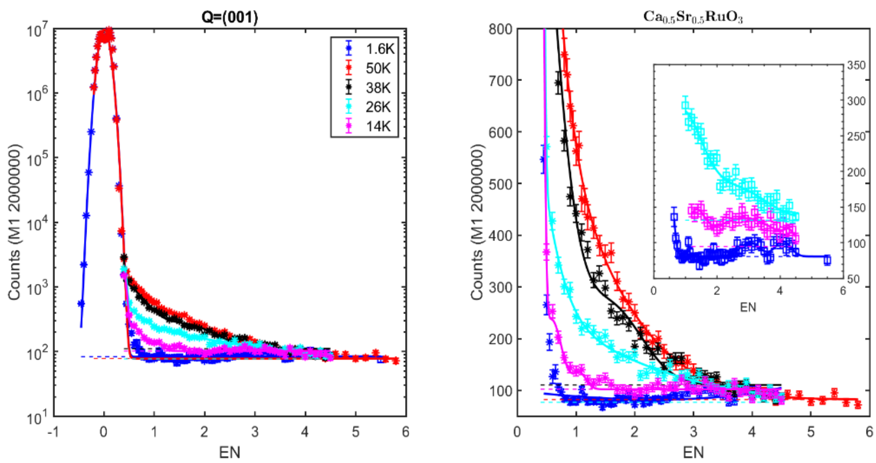

Fig. 3 Constant-***Q*** scans at the ferromagnetic zone center (100) for 50% of Ca doping. At low temperature one distinguishes two gaps at even higher energies that decrease upon heating. But there is clear evidence for excitations below these gaps at all temperatures.

For x=0.5, the low-temperature constant energy scans yield more stringent evidence for finite signals appearing below the gaps that we tentatively attribute to longitudinal excitations. Between 1.5 and 3.5 meV, there is a single peak visible, whose width does not change with energy. In contrast, the data at 50 K indicate additional broadening to emerge already at lower energies. Above 4 meV we find the expected split signal illustrating the magnon dispersion.

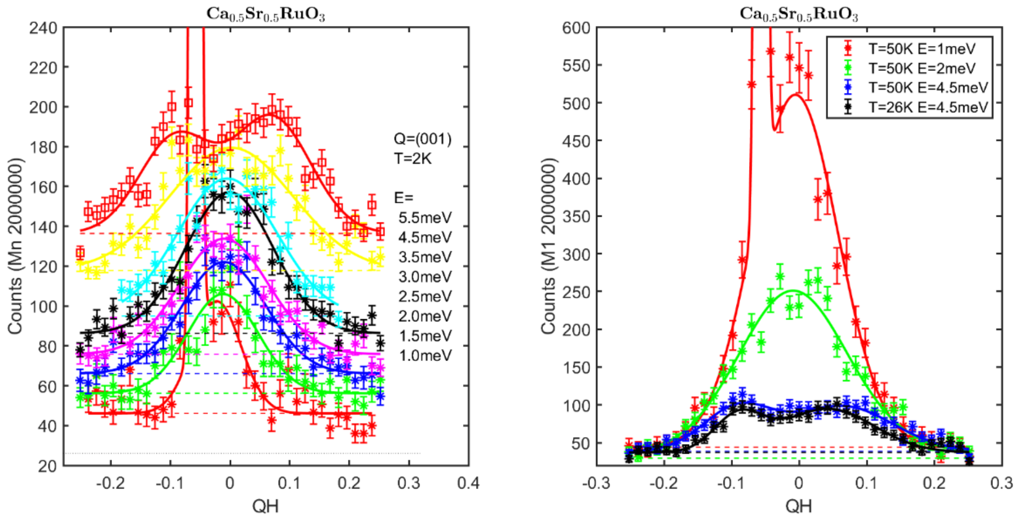

Fig. 4: Constant-energy scans across the magnon and paramagnon dispersion in $Sr_{0.5}Ca_{0.5}RuO_3$. While at low temperature the magnon dispersion only starts above the further enhanced gap values, separated modes already appear at much lower energy at higher temperature.

In summary, the inelastic neutron scattering experiments on two substituted $Sr_{1-x}Ca_xRuO_3$ with x=0.3 and 0.5 revealed some surprising results. First the anisotropy gaps are not following the trend of suppressed magnetic moments and reduced Curie temperatures but are strongly enhanced by more than a factor 2.5 already for x=0.3 and even more strongly for x=0.5. While the strange temperature dependence of the gap in $SrRuO_3$ let one expect some anomalous behavior also for the substituted materials, the remarkable enhancement of the anisotropy gaps indicates an enhanced impact of spin-orbit coupling that must rely on some new mechanism. Second, there is evidence for magnetic signal appearing below the gap that we tentatively attribute to longitudinal fluctuations.

References

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