EUROPEAN SYNCHROTRON RADIATION FACILITY

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# Experiment Report Form

**The double page inside this form is to be filled in by all users or groups of users who have had access to beam time for measurements at the ESRF**.

Once completed, the report should be submitted electronically to the User Office using the **Electronic Report Submission Application:**

*http://193.49.43.2:8080/smis/servlet/UserUtils?start*

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Reports can now be submitted independently of new proposals – it is necessary simply to indicate the number of the report(s) supporting a new proposal on the proposal form.

The Review Committees reserve the right to reject new proposals from groups who have not reported on the use of beam time allocated previously.

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All users must give proper credit to ESRF staff members and proper mention to ESRF facilities which were essential for the results described in any ensuing publication. Further, they are obliged to send to the Joint ESRF/ ILL library the complete reference and the abstract of all papers appearing in print, and resulting from the use of the ESRF.

Should you wish to make more general comments on the experiment, please note them on the User Evaluation Form, and send both the Report and the Evaluation Form to the User Office.

## Deadlines for submission of Experimental Reports

- 1st March for experiments carried out up until June of the previous year;
- 1st September for experiments carried out up until January of the same year.

## Instructions for preparing your Report

- fill in a separate form for each project or series of measurements.
- type your report, in English.
- include the reference number of the proposal to which the report refers.
- make sure that the text, tables and figures fit into the space available.
- if your work is published or is in press, you may prefer to paste in the abstract, and add full reference details. If the abstract is in a language other than English, please include an English translation.

| ESRF | **Experiment title:** Role of spin-orbit coupling in $Ca_{2-x}Sr_xRuO_4$ | **Experiment number**: HE 2823 |
|---|---|---|
| **Beamline**: ID08 | **Date of experiment**: from: 27 Nov. 2008 to: 02 Dec. 2008 | **Date of report**: |
| **Shifts:** 15 | **Local contact(s)**: Nicholas Brookes ( email: brookes@esrf.fr ) | *Received at ESRF:* |

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## Report:

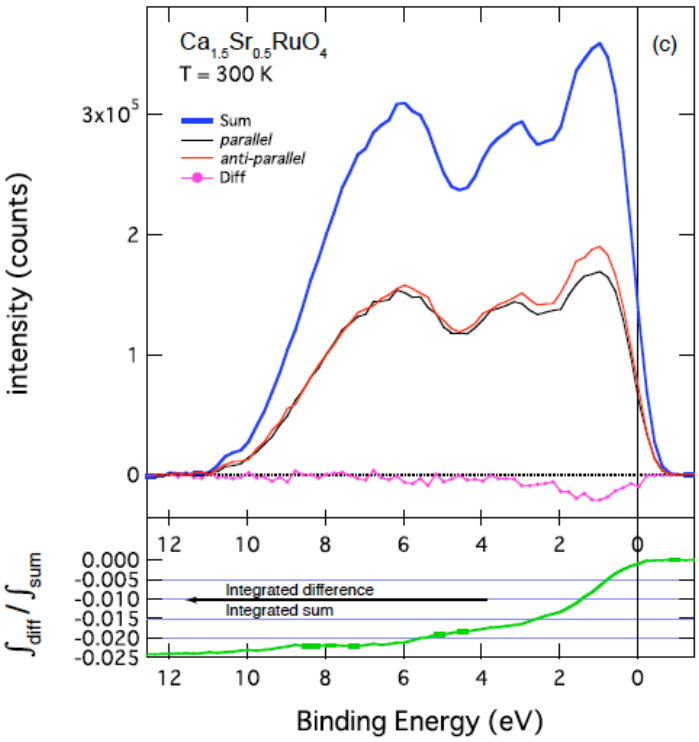
The layered and quasi two-dimensional $Ca_{2-x}Sr_xRuO_4$ system shows a very rich phase diagram as a function of the *x*-composition, temperature and magnetic field [1]. The end member $Ca_2RuO_4$ is an antiferromagnetic Mott-insulator with a Néel temperature of $T_N$ = 110 K, and shows a first order structural phase transition to a metallic state at $T_{MIT}$~ 350 K. In substituting Sr for Ca in the concentration range of $0.05 < x < 0.2$, the $T_{MIT}$ is lowered and becomes equal to $T_N$. In the range of 0.2<x<0.5, metamagnetism has been reported, and the $x = 0.5$ compound is considered to be a quantum critical point between a magnetic spin fluctuation state and a paramagnetic state. The anomalous mass-enhancement and ferromagnetic instability below 5 K are also reported for $x$~0.5. The other end member, $Sr_2RuO_4$, is an unconventional metal and turns out to be a *p*-wave superconductor below $T_C$ = 1.5 K.

Previous spin-resolved circularly polarized photoemission on $Ca_2RuO_4$ has revealed the presence of a significant spin-orbit signal in the Ru 4*d* shell [2]. This finding could be of immediate relevance for the question whether the ruthenates are the ideal system for the study of the orbital-selective Mott-transition, since the spin-orbit interaction will couple the different local Ru 4*d* $t_{2g}$ orbital and spin-quantum numbers as to produce bands with very mixed characters. The finding also raises directly the question whether this is also true for the metallic compositions, in particular $Sr_2RuO_4$. In fact, it is theoretically predicted very recently that the spin-orbit interaction does play a crucial role in determining the Fermi surface [3], very much in contrast to what is widely believed so far. And most interestingly, it is known in inorganic chemistry that isolated $Ru^{4+}$ ions tend to be non-magnetic [4], which reflects an atomic multiplet effect of the spin-orbit coupling, which requires a multi-Slater determinant explanation going beyond standard band structure approaches. This aspect may in fact form an unconventional starting point to model the occurrence of metamagnetism for 0.2<x<0.5. It is therefore essential to determine experimentally the magnitude of the spin-orbit signal in the $Ca_{2-x}Sr_xRuO_4$ system with varying Sr concentrations in order to

investigate how much the spin-orbit signal changes in going through the MIT, since this may elucidate directly how the local spin-orbit physics competes with band formation.

We have carried out the *spin-resolved* photoemission measurements using *circularly polarized* light on $Ca_{2-x}Sr_xRuO_4$ single crystals, 0<x<0.5 and x= 2, in order to reveal the role of the spin-orbit interaction for the electronic structure of the ruthenate system. This type of photoemission has been developed only recently with very promising results for determining the spin-orbit quantum numbers and orbital momentum in transition metals and oxides [5]. Magneto-optical sum rules are available with which the expectation value $\langle \Sigma_i l_{x',i} s_{x,i} \rangle$ can be determined directly from the experiment without need for detailed simulations of the spectral line shape [6]. Nevertheless, the low-count rates associated with the low efficiency of standard spin detectors have hampered a wider application of this method. For this ruthenate project in which the interest is in sum-rule numbers, we utilized a state-of-art Time-of-Flight spin-detector (Mott-TOF) [15] recently built by the group of N.B. Brookes at ID08. This new detector - for sum-rule experiments - yields a gain in performance rates of at least factor 5 in comparison to other detectors presently available in the world.

Figure 1 shows the valence band spectra of $Ca_{1.5}Sr_{0.5}RuO_4$ obtained by the spin-resolved circular polarized photoemission with 700 eV-photons at 300 K. One can observe a clear difference between the spectra taken with the photon spin (given by the helicity of the light) parallel (black line) or antiparallel (red line) to the electron spin. The relevant quantity to be evaluated here is the integrated intensity of the difference spectrum (maganta lin) relative to that of the integrated intensity of the sum spectrum (blue line). The preliminary analysis gives the $\int$ *diff* / $\int$ *sum* number with -0.024 ±0.004. This number is quite sizable as compared with $Ca_2RuO_4$, which has the $\int$ *diff* / $\int$ *sum* number with -0.041 ±0.011 at 150 K [2]. Since the expectation value $\langle \Sigma_i l_{x',i} s_{x,i} \rangle$ is given in proportion to the $\int$ *diff* / $\int$ *sum* number, this gives evidence that the spin-orbit interaction must be considered seriously for the understanding of the magnetic properties of this composition.




Using a local cluster *ansatz*, we find that the ground state of the $Ru^{4+}$ ion is a singlet, i.e. non-magnetic, if we take the spin-orbit interaction into account. This means that we now have another starting point for the mechanism of metamagnetism in this compound: the Ru ion likes to be in a non-magnetic state as far as the local physics are concerned and we have to add band formation in order to drive the system into a magnetically active system.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