Searches for the spatial nature of magnetic order in $Sr_{1.5}Ca_{0.5}RuO_4$

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One of the crowning achievements of condensed matter physics in the last century was a theory by Bardeen, Cooper and Schrieffer ("BCS") explaining the mechanism of superconductivity (SC) in certain metals, the abrupt decrease of a material's electrical resistivity to zero below a critical temperature T_C, as a result of interactions between conduction electrons and phonons, vibrations of the crystalline lattice.

However, in the last three decades a variety of superconductors whose properties deviate significantly from BCS predictions have been discovered, among them the famous high-T_C cuprate and iron-based superconductors. Despite 30 years of intensive research, a comprehensive theory of such "unconventional" superconductivity remains elusive. However, it is clear that magnetic interactions play a key role in the development of SC in these materials. In particular, in a wide variety of superconducting families static magnetic order exists as a competing state very near to superconductivity. To that end, careful magnetic characterization of superconductors and related compounds produced by chemical substitution or applied pressure are crucial in order to elucidate the mechanism by which SC emerges from a competing magnetically ordered state.

One such system is the layered perovskite Sr_2RuO_4, discovered to be a superconductor with $T_C = 1.5K$ [1]. Static magnetic order was found in the isostructural but non-superconducting Ca_2RuO_4, as well as in doped compounds produced by small Sr substitution on the Ca sites ($Ca_{2-x}Sr_xRuO_4$ with $x \leq 0.5$) or partial replacement of Ru^{4+} ions with Ti^{4+} ($Sr_2Ru_{1-y}Ti_yO_4$ with $y \geq 0.025$) [2,3], although the relationship between SC in Sr_2RuO_4 and magnetism in these doped compounds remained unclear. Recently we have found [4] using muon spin relaxation (μSR) and susceptibility that static magnetic order exists throughout almost the entire substitutional space of $Ca_{2-x}Sr_xRuO_4$, aside from a narrow region surrounding the superconducting Sr_2RuO_4 (Fig. 1), demonstrating that magnetic order exists as a competing state very near to SC order, and reinforcing this intriguing commonality between various families of unconventional superconductors.

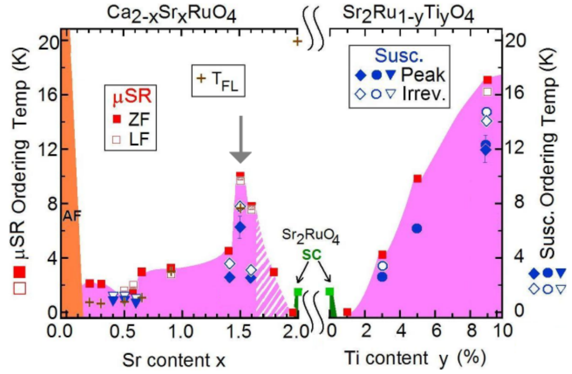

Fig. 1 Magnetic phase diagram of $(Ca,Sr)_2RuO_4$ and $Sr_2(Ru,Ti)O_4$ determined g zero-field (ZF) and longitudinal-field (LF) μSR and magnetic susceptibility, demonstrating the existence of magnetic order (violet shading) throughout most of the substitution space between antiferromagnetic (AF) Ca_2RuO_4 and superconducting (SC) Sr_2RuO_4. The sample of present interest, $Ca_{0.5}Sr_{1.5}RuO_4$, is arrowed. From Ref. [4].

Although μSR and susceptibility measurements are highly sensitive to the existence of magnetic order, neither is able to definitively decipher its long-range nature. In $Ca_{0.5}Sr_{1.5}RuO_4$, for example (arrowed in Fig. 1), our μSR measurements found static magnetic order below T = 10 K, and fits to the susceptibility data yielded a negative Weiss temperature indicative of antiferromagnetic (AF) correlations. The time dependence of the muon spin polarization, and the presence of a peak in ac susceptibility Im(χ_{ac}), are suggestive of spin-glass behaviour, but a direct determination of spatial correlations can only be made with a spatially sensitive probe.

Neutron scattering is a reciprocal-space probe uniquely suited to magnetic structure determination. A neutron scattering experiment performed on $Sr_2Ru_{0.91}Ti_{0.09}O_4$ [5] found that a short-range incommensurate AF state sets in at $T_N \approx 25K$, with an average ordered moment of $0.3\mu_B$

per Ru site. Based on our μSR results on $Ca_{0.5}Sr_{1.5}RuO_4$, we would expect to find magnetic order below T ≈ 10K, with a $0.2\mu_B$ effective ordered moment per Ru site.

To further elucidate the spatial nature of this order, we conducted measurements using the CNBC C5 and D3 spectrometers on a small single-crystal specimen of $Ca_{0.5}Sr_{1.5}RuO_4$. Due to the small available sample size ($1.5x3x7mm^3$, 130 mg) and the small ordered moment, any magnetic response was expected to be weak. Further, since the magnetic structure is expected to be 2-D with short correlation lengths, any intensity would be broadly distributed in reciprocal space. Hence, we optimized our configurations to maximize the signal and the signal-to-noise ratio. We used pyrolytic graphite (PG) monochromators and analyzers at both instruments in order to maximize the flux at the sample and minimize background at the detector; at D3 the availability of a tall PG monochromator made it possible to deliver additional beam intensity to the sample. Using two PG filters with λ = 2.37 Å neutrons minimized λ/2 contributions to the signal. Due to the small sample dimensions, the slits were placed immediately next to the cryostat and closed down to the maximum extent possible to reduce background.

High-statistics scans were performed along high-symmetry directions and around Fermi-surface nesting positions that have yielded magnetic responses in inelastic studies of Sr_2RuO_4, and diffraction measurements on $Sr_2Ru_{0.91}Ti_{0.09}O_4$, although no differences were seen between scans above and below the expected transition temperature. A mesh scan was performed over a wide swath of the HK0 plane (Fig. 2), although no statistically significant differences between high and low temperature were found.

This failure to detect a magnetic signal could be consistent with intrinsic spin-glass behaviour in $Ca_{0.5}Sr_{1.5}RuO_4$, as may be inferred from the μSR and ac susceptibility results. However, given the small sample size and small ordered moment, these results could also be consistent with incommensurate magnetic order as is seen in $Sr_2Ru_{0.91}Ti_{0.09}O_4$, if accompanied by very short correlation lengths.

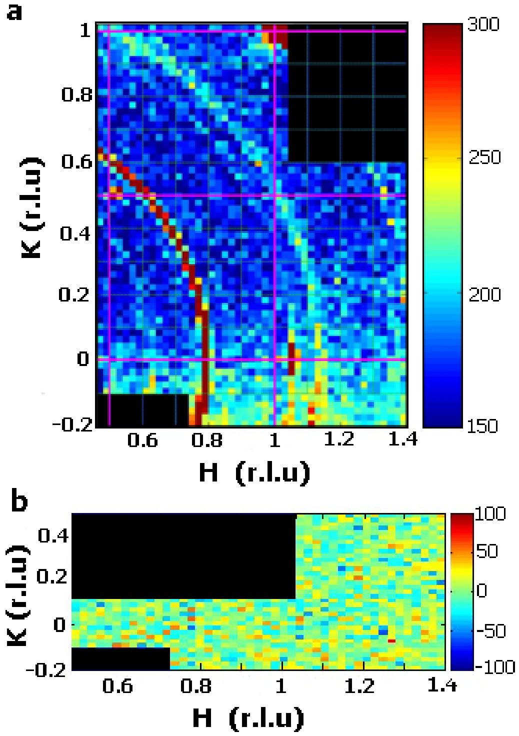

Fig. 2 (a) C5 [HK0] plane mesh scan at T = 3 K. The rings are due to the Al sample holder, and the [110] nuclear Bragg peak (and λ/2 contributions) can be seen. (b) Scattering difference between T = 3 K and 50 K. No statistically significant differences between the scattering intensities are observed.

References

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