Magnetic critical scattering from the spin-orbit Mott insulator Sr$_2$IrO$_4$

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The group of 5d transition metal oxides (TMOs) exhibits a wide range of interesting physical phenomena, due to the spin-orbit coupling of the 5d ions being a significant factor in the electronic and magnetic behaviour of systems in this group. Particular interest lies in so-called $J_{\text{eff}} = \frac{1}{2}$ systems, formed as a consequence of the subtle interplay between the cubic crystal field and intrinsic electron correlation. [1,2] Magnetic critical scattering is a useful technique with which to probe the critical dynamics of a system close to the magnetic transition temperature, $T_c$, and has been used to study a number of 3d TMOs [3]. Using synchrotron X-ray radiation, it is possible to determine the wavevector and temperature dependence of the critical scattering to very high resolution. In particular, X-ray critical scattering is particularly sensitive to subtle changes in the shape of the resultant scattering function, which is key to determining the relative length scales of correlation between regions of spins.

The iridium perovskite compound, Sr$_2$IrO$_4$, has been implicated as a possible candidate for exhibiting 2D Heisenberg antiferromagnetic behaviour. In Sr$_2$IrO$_4$, the iridium magnetic moments are ordered in the ab-plane below the Néel temperature, $T_N$. Above $T_N$ however, Fujiyama and colleagues [4] showed via magnetic critical scattering that the in-plane correlation length appeared to follow the theoretical formula by Makivic [5] for a 2D Heisenberg antiferromagnet, with a nearest neighbour exchange constant $J = 100 \pm 10$ meV. However this value does not agree with values obtained via resonant inelastic X-ray scattering (RIXS) measurements and theory calculations. Measurements had previously been carried out on this material at Diamond, which suggested that the high temperature limiting behaviour was consistent with a 2D Heisenberg model. However there was insufficient data at high temperature to confirm this hypothesis. The experiment at P09 was performed predominantly in order to obtain this missing data.

Resonant magnetic scattering data was obtained at P09 on a sample of Sr$_2$IrO$_4$ (approximate dimensions 2 x 1 x 0.1mm). In order to maximise the magnetic contribution to the scattering, the incident energy was chosen to correspond to the Ir L$_3$ edge at 11.214 keV. This value was chosen based on the maximum of the iridium fluorescence. The incident beam was focussed using 64 compound refractive lenses and an exit slit size of 2 x 2 mm, which gave an estimated beam size of 60 x 60 um. A gold (3 3 3) polarization analyser crystal was used to discriminate between polarization channels and suppress the fluorescent background.

Unfortunately, there were a number of issues with the experiment which meant that the data that was collected was severely limited. Firstly the scheduling of only 12 shifts as opposed to the 18 that are usually required meant that the experiment was always going to be time-constrained. A further impact was caused by the late finish of the previous experiment, followed by a change of hutch between experiments. This meant around three hours were lost from changing configuration. The main problem however was a lack of temperature stability for a large proportion of the experiment, a key requirement for accurate critical scattering measurements. Measurements were initially taken
with the cryofurnace, however it was found that intensity persistently dropped off and came back with time. It was found that the drop in intensity was correlated with a change in temperature (Fig. 1a), which suggested an issue with the cryofurnace or the temperature controller. Adjusting PID parameters had little effect, so the cryofurnace was switched with a 4K Displex and an alternative controller. Unfortunately a similar effect occurred with the new setup, with typical oscillations of ±1K. This was attributed to a faulty Lakeshore, so finally the 4K Displex was used with the original Lakeshore. Temperature stability was finally achieved at lunchtime on the third day, meaning only 19 hours of meaningful measurements could be performed out of a possible 96 hours.

The limited measuring time meant that only a small amount data could be obtained. Nevertheless, the experimental results agree quite well with the results that were obtained at Diamond (Fig. 1b). The correlation length out-of-plane decays much faster than that in-plane, with the high temperature limiting behaviour appearing to be that of a 2D Heisenberg antiferromagnet. This is consistent with previous measurements. The signal-to-noise ratio at P09 however was considerably worse than that at Diamond, and counting times of 3 to 4 times longer were required to obtain comparable statistics. The addition of compound refractive lenses has the advantage of reducing the beam size significantly, at the expense of incident flux. This may be the reason for the lack of signal. Without the problems at the beginning of the experiment, it should have been possible to compensate for this. More measurements have been performed in the meantime and a paper is currently in preparation.

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References