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Experimental report

Proposal code: HC-2253
Proposal title: Paramagnons in the pseudogap phase of electron doped $(Sr_{1-x}La_x)_2IrO_4$

Abstract
We report on a novel resonant inelastic x-ray scattering (RIXS) study of the collective magnetic excitations in both parent Sr_2IrO_4 and its electron doped version $(Sr_{1-x}La_x)_2IrO_4$. A sizeable gap in the excitations spectrum at all doping levels is reported here for the first time, which arises from a significant anisotropy in the interaction between pseudo-spins. The results are interpreted in terms of a 2D XYh_4 magnetic Hamiltonian and the magnitude of the anisotropies is extracted by a fit of the model to the measured dispersion.

Experimental method
The RIXS measurements were performed at the ID20 beamline at the European Synchrotron Radiation Facility (Grenoble, France). The experiment was carried out in horizontal scattering geometry using a spherical ($R = 2\ m$) Si(844) diced analyzer with a $60\ mm$ mask and a Si(844) secondary monochromator. This gives an overall energy resolution of $FWHM = 23.41\ meV$ and an in-plane momentum resolution of $\Delta Q_\perp = 0.1\pi$. The samples were cooled down below the Néel transition ($T_N \approx\ 240K$) by means of a He-flow cryostat.

Samples
Single crystals of $(Sr_{1-x}La_x)_2IrO_4$ with varying La concentration ($x = 0, 0.01, 0.04$) were flux grown by standard methods and characterized by resistivity and susceptibility measurements. The doping level of each of the crystals used for the present investigation was checked by means of energy dispersive x-ray spectroscopy (EDX). The doping dependence of the magnetic correlation length was also checked through reciprocal space scans of magnetic diffraction peaks performed using the RIXS spectrometer: consistently with previous reports [1,2], long-range order is suppressed in the doped samples while short-range correlations persist in the basal plane of the crystal. Substitution of trivalent La for divalent Sr dopes the system with electrons while preserving the $I4_1/acd$ crystal structure and the strong spin-orbit coupling of the Ir atom [2]. The momentum transfer values $Q = (Q_x\ , Q_y\ , Q_z)$ mentioned in this report are quoted in units of $1/a$ for their in plane component $Q_\perp = (Q_x\ , Q_y)$, where $a = 3.89$ Å is the in-plane lattice constant of the undistorted $I4/mmm$ unit cell, and the miller index L (which was kept fixed to $L = 33$ throughout the whole experiment) for their out-of-plane part Q_z.

Description of the measurements and results summary
RIXS spectra were collected at $T = 20\ K$ keeping the incident energy fixed to the Ir L_3 absorption edge ($E = 11.215\ keV$) and measuring the scattered photons energy in the energy loss range $E_{loss} = -0.2 \div 0.6\ eV$. For each value x of La content, several spectra were collected for different values of Q along high symmetry directions of the (0,0,33) first Brillouin zone (BZ) (chosen for providing a scattering angle $2\theta \approx 90°$ and thus suppressing charge elastic scattering). These are plotted in the intensity maps of Fig. 1(a-c). Longer spectra up to $E_{loss} = 1.2\ eV$ were collected for the high symmetry points $Q_\perp = (0,0)\ ,(\pi,\pi)\ ,(0,\pi)\ ,(\pi/2,\pi/2)$ (Fig.1(d)). As first reported by Kim et al [3], the parent compound data show a collective magnetic excitation dispersing from the AF propagation vector (π,π) and extending up to about $0.2\ eV$ and a spin-orbit exciton around $0.6\ eV$ arising from intra-t_{2g} transitions. The doped compounds display analogous energy loss features. In particular, heavily damped magnetic excitations

with a similar in-plane dispersion survive up to highest doping level, where the long range magnetic order is suppressed. These findings qualitatively agree with the results from previous studies [1,4] and further strengthen the link to the hole-doped cuprates, where spin-wave like excitations (paramagnons) are present in the metallic and superconducting regions of the phase diagram [5].

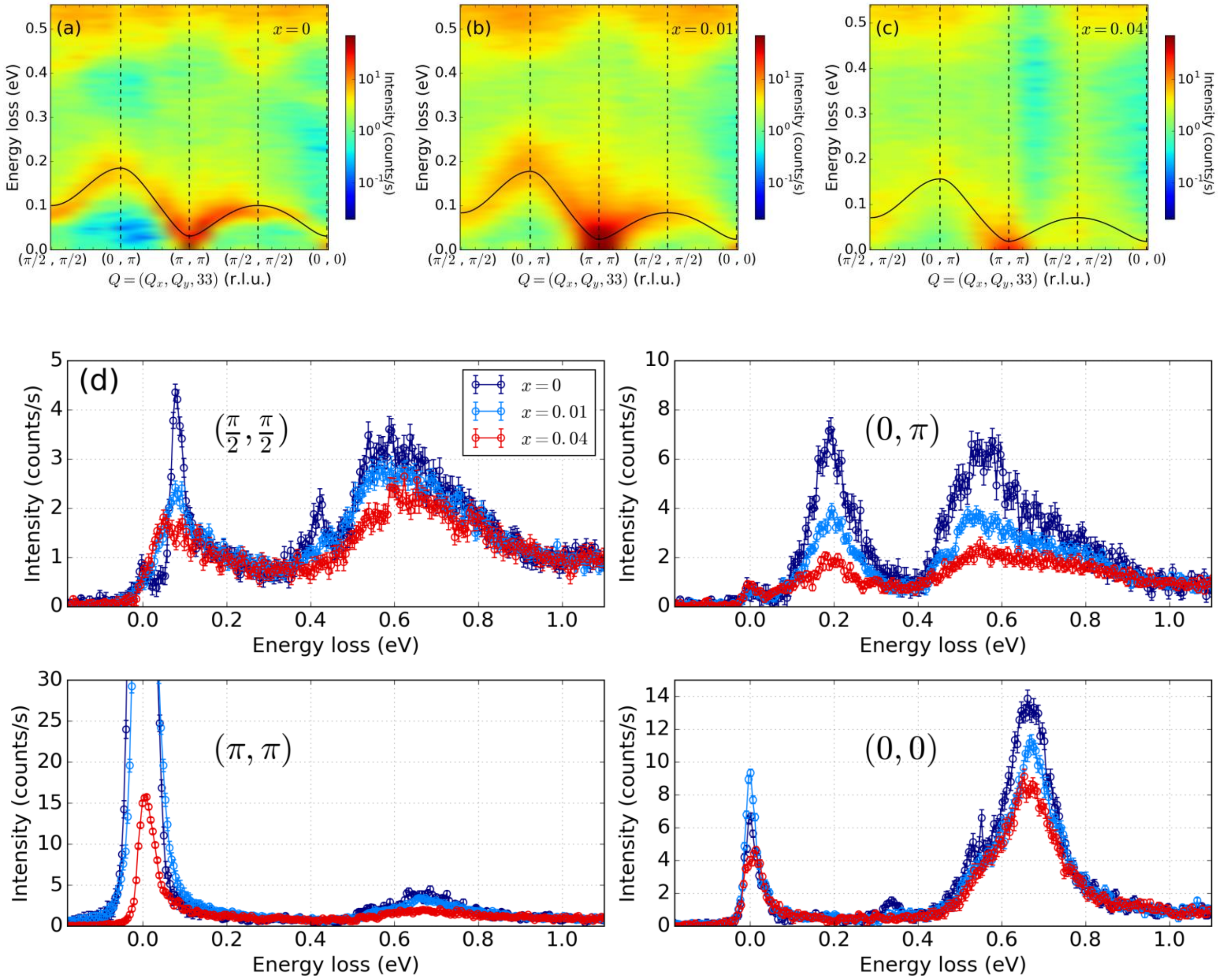

Figure 1. RIXS spectra as a function of the in-plane momentum transfer Q along high-symmetry directions of the first Brillouin zone (BZ) for $x = 0$ (a), $x = 0.01$ (b) and $x = 0.04$ (c). The inset in (a) shows the $I4/mmm$ (large square) and $I4_1/acd$ (small rotated square) BZ and the path along which the spectra were collected. The solid line represents the best fit of the single magnon energy to the XYh_4 model discussed in the text. The spectra corresponding to the high symmetry Q values marked by the vertical dashed lines are plotted in (d) as a function of the doping level. In (d) the spectra of different samples for each Q point were normalized using the spectral weight around $E_{loss} = 1$ eV.

The novel feature emerging from the data presented in this report is the presence of a finite energy gap in the magnetic excitations spectrum. This is evident from the low energy detail of the spectra collected at the crystallographic zone center and the AF propagation vector shown in Fig. 2(a) and 2(b), respectively. Here, the spectra for different doping levels are plotted together with the fitted single magnon profile represented by the shaded area. As it is evident already by simple inspection of the raw data, magnetic excitations with a non-vanishing energy are clearly resolved as a separate energy loss peak residing close to (and partially overlapping with) the elastic line. The magnitude of the gap is about $23\,meV$ (roughly the same at (0,0) and (π,π)), with no significant doping dependence. Another salient feature emerging from inspection of Fig. 2(a-b) is the fact that, as well as displaying a doping-independent non-zero energy, the magnon peak at (0,0) and (π,π) does not considerably broadens as the dopant concentration is increased. For other values

of the momentum transfer however (Fig. 2(c-d)), the response of the magnon upon injection of free carriers is markedly different, revealing both a significant softening and damping for increasing x values.

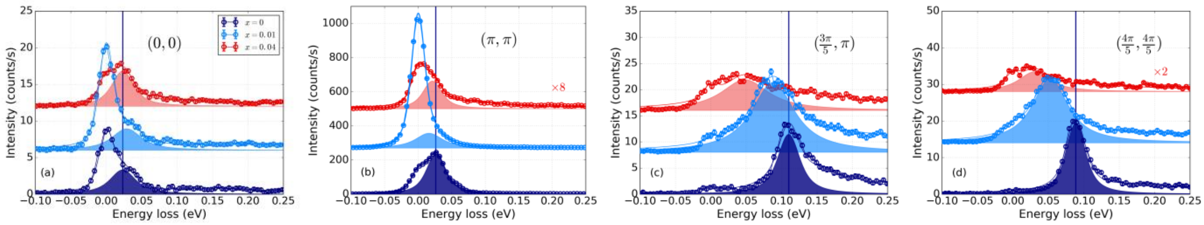

Figure 2. Doping level dependence of the low energy region of the RIXS spectra for different values of the in-plane momentum transfers. The errorbars represent the measured intensity analogously to Fig. 1(d) while the solid line refer to the fit of the spectrum as described in the text. The shaded regions show the fitted single magnon peak, highlighting the presence of a finite gap at (0,0) and (π, π). The vertical solid line marks the fitted magnon energy in the parent compound ($x = 0$).

The observed gapped magnon dispersion can be accounted for by a XYh_4 magnetic Hamiltonian:

$$H = \sum_{\langle i,j \rangle} J_1 \vec{S}_i \cdot \vec{S}_j + \sum_{\langle\langle i,j \rangle\rangle} J_2 \vec{S}_i \cdot \vec{S}_j + \sum_{\langle\langle\langle i,j \rangle\rangle\rangle} J_3 \vec{S}_i \cdot \vec{S}_j + D \sum_i (S_i^z)^2 + \frac{1}{2} e \left(S_i^{+4} + S_i^{-4} \right) \qquad (1)$$

where $\vec{S}_i$ is the i-th Ir^{4+} pseudo-spin. The first three terms correspond to a traditional Heisenberg model with nearest neighbours (n.n.), next n.n. and next next n.n. interactions (J_1, J_2 and J_3 are the corresponding exchange integrals). The easy-plane anisotropy responsible for confining the moments in the IrO_2 planes is modelled by D, while the four-fold term e (also referred to as basal plane anisotropy) breaks the symmetry within the plane. The gap at all doping levels is correctly reproduced by a value of the easy-plane anisotropy D ranging from 4.2 meV for the undoped compound down to 2.0 meV for the highest doping level (see solid lines in Fig. 1(a-c)). Though smaller, these values are not negligible when compared to the isotropic exchange integrals J_i. The origin of the in-plane anisotropy can be attributed to the sizeable tetragonal distortion of the IrO_6 octahedra away from the perfect cubic symmetry of the $J_{eff} = 1/2$ state found in Sr_2IrO_4 [6]. In particular, the values for D found in the present investigation are consistent in the order of magnitude with the tetragonal field $|\Delta| = 10\ meV$ measured by resonant elastic scattering [7] on the undoped compound and with the upper limit of 190 meV estimated by [6]. A significant easy-plane anisotropy in the pseudo-spins interaction also confirms the attribution [8] of the critical behaviour of the Sr_2IrO_4 order parameter to the XYh_4 universality class. As for the basal plane anisotropy, our choice of fixing $e = 0$ is justified by the fact that this term is expected to be negligible [9] with respect to the in-plane one.

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