ESRF	**Experiment title:** Role of spin-orbit coupling in correlated 3*d*-5*d* double perovskites La_2MnReO_6 and La_2NiReO_6	**Experiment number**: HC-2071
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Report:

Double perovskites are multi-cation compounds of the form $A_2BB'O_6$ with a rare earth or an alkaline earth element at the *A*-site and a combination of 3*d*-3*d*, 3*d*-4*d*, or 3*d*-5*d* transition metals at the *B*/*B*' sites. Double perovskites offer the possibility to engineer a wide range of physical functionalities such as half-metallicity, high-T_c magnetism, and multiferroicity. In particular, the 3*d*-5*d* based compounds have attracted an increased scientific interest due to strong correlation effects in combination with enhanced spin-orbit coupling.

We synthesized thin films of the metastable double perovskites La_2MnReO_6 (LMRO) and La_2NiReO_6 (LNRO) using pulsed laser deposition. Density functional theory calculations performed by the Ležaić-group suggested, that both compounds are ferrimagnetic insulators exhibiting a strong spin-orbit coupling effect of opposite sign. The integral magnetic moments of LMRO and LNRO thin films were characterized using a superconducting quantum interference device (SQUID). The magnetization *vs.* field curves for both compounds show well defined hysteresis loops with different anisotropies (Fig. 1). The observed saturation magnetizations of 2.3 ± 0.1 and 0.4 ± 0.1 μ_B/f.u. for LMRO and LNRO, respectively, are in good agreement with the theoretically predicted ferrimagnetic order and indicate the presence of a significant orbital contribution. In order to characterize the contribution of spin-orbit coupling and to compare the role of Re magnetism in the two compounds, X-ray magnetic circular dichroism (XMCD) measurements at Re $L_{2,3}$ X-ray absorption edges were performed in this experiment.

Absorption spectra were recorded using the total fluorescence yield detection mode. The XMCD spectra for both LMRO and LNRO were obtained as direct difference between consecutive XANES scans (X-ray Absorption Near Edge Spectrum) recorded with opposite helicities of the incoming X-ray beam. To ensure artefact-free measurements, the data were collected for two opposite directions of the applied magnetic field of 2 T (parallel and antiparallel to the X-ray beam). All measurements were performed at 20 K which is well below the observed T_c for both LMRO and LNRO.

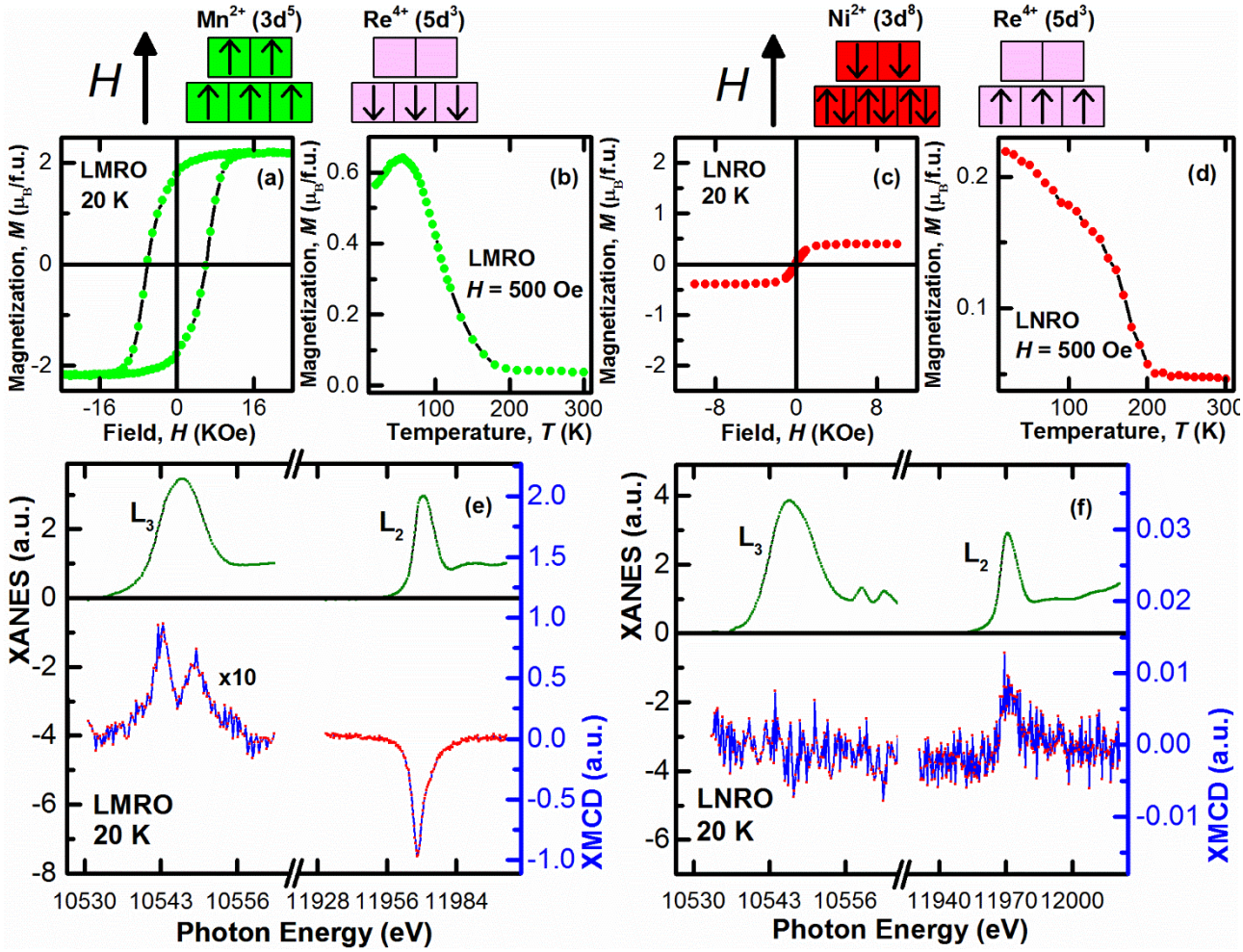

Fig. 1. Magnetization *vs.* field and field cooled magnetization *vs.* temperature curves for (a,b) LMRO and (c,d) LNRO thin films measured with a SQUID magnetometer. XANES and XMCD spectra of the Re $L_{2,3}$ edges for (e) LMRO and (f) LNRO thin films measured at 20 K. The *d*-shell electron configurations of the $3d^5$-$5d^3$ (Mn^{2+}-Re^{4+}) and $3d^8$-$5d^3$ (Ni^{2+}-Re^{4+}) cation pairs as per the theoretically expected ferrimagnetic order are schematically shown at the top for LMRO (left) and LNRO (right), respectively.

The measured XANES spectra of LMRO and LNRO are shown in Fig. 1(e) and (f). In the case of LMRO, a clear XMCD signal indicates a significant magnetic moment at the Re-site (Fig. 1 (e)). The shape of the LMRO XMCD spectrum shows that the overall Re moment is aligned opposite to the net magnetization, which is consistent with theory. Furthermore, using the sum rules, we estimated a large 5*d* spin moment at the Re-site, m_S = -1.15μ_B and a significant orbital moment, m_L = 0.33μ_B (resulting $|m_L/m_S|$ = 0.29). These values are in fair agreement to the Re moments obtained from the band structure calculations for LMRO (m_S = -0.96μ_B, m_L = 0.30μ_B and $|m_L/m_S|$ = 0.31).

The LNRO XMCD signal at the Re L_2 edge is aligned with the external magnetic field and opposite in sign to that of LMRO. This is a fingerprint of the oppositely aligned Re moments in these two compounds with respect to the external magnetic field. For LNRO, the magnitude of the XMCD signals is an order of magnitude smaller than for LMRO (Fig. 1(f)). The small film thickness and artefacts such as the Bragg reflections from the $SrTiO_3$ substrate limited the resolution of the LNRO XMCD spectrum and restricted a quantitiative analysis via sum rules.

In summary, based on our XMCD measurements, we confirm the presence of an induced moment at the Re-site in both LMRO and LNRO. Quantiative estimations of spin and orbital moment in the case of LMRO confirm a large unquenched orbital moment at the Re-site, which contributes to the observed total magnetic moment. Furthermore, by comparing the qualitative features of the spectra for both LMRO and LNRO, the overall moments at the Re-site were found to be aligned oppositely in the two cases as per the theoretical predictions.

Two papers on synthesis, structural and magnetic properties of LMRO and LNRO including the results obtained from the XMCD measurements are in preparation.