Dynamical local correlations in Magnetite revealed by Fe 2p3d RIXS MCD

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Introduction

Magnetite (Fe_3O_4) is an archetype correlated system where the charge, orbital and spin degrees of freedom compete. Although widely studied, the ground state of Fe above and below the Verwey transition are intensely debated. The low temperature phase was proposed to emerge as a result of the condensation of linear orbital molecules - the trimeron- promoted by local electron interactions [1]. Diffuse X-ray scattering experiments indeed suggested the existence of dynamic polarons in the high temperature phase that self-organises at low temperatures, however the ordering pattern was deduced to be much more complex than the linear trimeron [2]. In order to shed light on the nature of the magnetic polaron, we measured for the first time high resolution Fe 2p3d RIXS magnetic circular and linear dichroism (MCD and MLD).

Methods

We measured high resolution Fe 2p3d RIXS (~85 meV) on a (001) Fe_3O_4 synthetic single crystal at the ADRESS beamline of SLS. A permanent magnet (~0.5T) was used to saturate the magnetic moments of Fe_3O_4 in two configurations; in and out of the scattering plane. The incident polarisation was rotated a full 360 degrees in addition to circular polarisation measurements.

Results and discussion

We observed a strong RIXS-MCD signal at the low loss energy feature (at 90meV). The magnetic response unambiguously identifies the excitation as a spin flip at the formal Fe^{3+} sites. The magnetic circular and linear dichroic behaviour of the d-d excitations confirms the existence of considerable local electron-electron interactions persisting above the Verwey transition inline with the idea of a dynamical magnetic polaron. Combining the experimental results with detailed multiplet ligand-field calculations using Wannier orbitals [3], we accurately quantified the local distortion parameters of the polaron. Fourier analysis of the angular dependences and phase shifts between the spectral features reveal that the local orbital moments of Fe in Fe_3O_4 are not fully quenched. Our results provide strong experimental evidence for noncollinear orbital ordering at the Fe sites. This explains the large discrepancies found in literature as regards to the orbital moment of Fe in Fe_3O_4 [4,5].

Conclusion

High resolution Fe 2p3d RIXS magnetic circular and linear dichroism measurements were performed on a Fe_3O_4 single crystal above the Verwey temperature. Strong magnetic dichroism enabled us to pinpoint the spin flip excitation and to accurately quantify the exchange interaction. The detailed study of the angular behaviour of the d-d excitations revealed the local distortion and orbital ordering of Fe in Fe_3O_4. Our results serve as a corner stone in our understanding of the interplay of the degrees of freedom in the Fe_3O_4.

References

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