Charge Transfer Properties of Zigzag-Antiferromagnet Nickel Phosphorus Trisulfide

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Abstract

Charge transfer insulators have been observed in several transition metal compounds before and has been distinguished from other phases such as the Mott-Hubbard insulator using the Zaanen-Sawatzky-Allen (ZSA) model considering the ligand-to-metal charge transfer energy (Δ) and the on-site Coulomb repulsion [1]. Metal phosphorus trichalcogenide (MPX_3) family was touted as a perfect magnetic 2D system due to the negligible direct exchange and extremely weak magnetic interactions across the layers [2]. A member of this family is nickel phosphorus trisulfide ($NiPS_3$) purporting properties likely of a charge transfer insulator. $NiPS_3$ is a zigzag-antiferromagnet having a Néel temperature of 155 K [3,4], with Ni atoms arranged in a honeycomb lattice with $(P_2S_6)^{4-}$ bipyramid perpendicular to this plane. In this work, we performed polarization-dependent x-ray absorption spectroscopy (XAS) to probe the electronic and magnetic properties of the material in both the paramagnetic and antiferromagnetic phase. With the conditions such as large electron affinity and low ligand electronegativity, $NiPS_3$ manifested a negative charge transfer value. The XAS result described the Ni^{2+} high spin state configuration with the holes in the d_{z^2} and $d_{x^2-y^2}$ orbitals. Starting with the previous calculations in $NiPS_3$[5], we performed charge transfer multiplet calculations [6] and the XAS feature related to Δ located around 3.3 eV above the L_3 peak. The position of the XAS charge transfer feature did not change as we go to the antiferromagnetic phase. Resonant inelastic x-ray scattering (RIXS) was performed for its sensitivity to probe the elementary excitations in $NiPS_3$[7] as well as to estimate the contribution of Δ to the ionic crystal field splitting e_g-t_{2g}.

References

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