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Discovery of Hidden Charge-Neutral Muon Centers in Magnetic Materials: Implications and Applications

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Spin polarized muons are widely known as an extremely sensitive local probe of magnetism. Additionally, positively charged muons implanted into semiconductors and insulators often bind an electron to form a charge-neutral muon-electron bound state frequently referred to as a muonium center. While studied extensively in non-magnetic semiconductors and insulators as light analogues of corresponding hydrogen centers, charge-neutral muon states are rarely considered relevant in magnetic materials. Apart from the singular exception of antiferromagnetic MnF_2 [1], no long-lived charge-neutral centers had been identified in magnetically ordered materials up-to-date.

Here, we present strong evidence that charge-neutral muon centers *do* exist in magnetic compounds. Detailed new μSR investigations of the antiferromagnets Cr_2O_3 [2], Fe_2O_3 [3] and MnF_2 , in conjunction with density-functional-theory calculations, reveal that charge-neutral muon states are present in magnetic materials and can form with different electronic structures, analogous to the variety of muonium centers found in non-magnetic materials.

Crucially, we find that in magnetic materials, charge-neutral muon states do not display any signatures conventionally associated with muonium centers, making it difficult to distinguish them from the often assumed positive charge state. We demonstrate that the presence of the additional charge alters the local electronic and magnetic structure, affecting the μSR signal and its relationship with the intrinsic magnetic properties. Since the muon is used extensively as a sensitive magnetic probe, it is imperative to understand under what conditions charge-neutral states are formed in magnetic materials, and what impact they have on the observed μSR frequencies and damping rates.

[1] Uemura *et al.*, *Hyperfine Interact.* **31** 313(1986)

[2] M.H. Dehn *et al.*, *Phys. Rev. X* **10**, 011036 (2020)

[3] M.H. Dehn, J.K. Shenton *et al.*, *Phys. Rev. Lett.* **126**, 037202 (2021)

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