Charge-neutral muon states in Cr_2O_3 and Fe_2O_3: investigating hydrogen impurities in transition metal oxides

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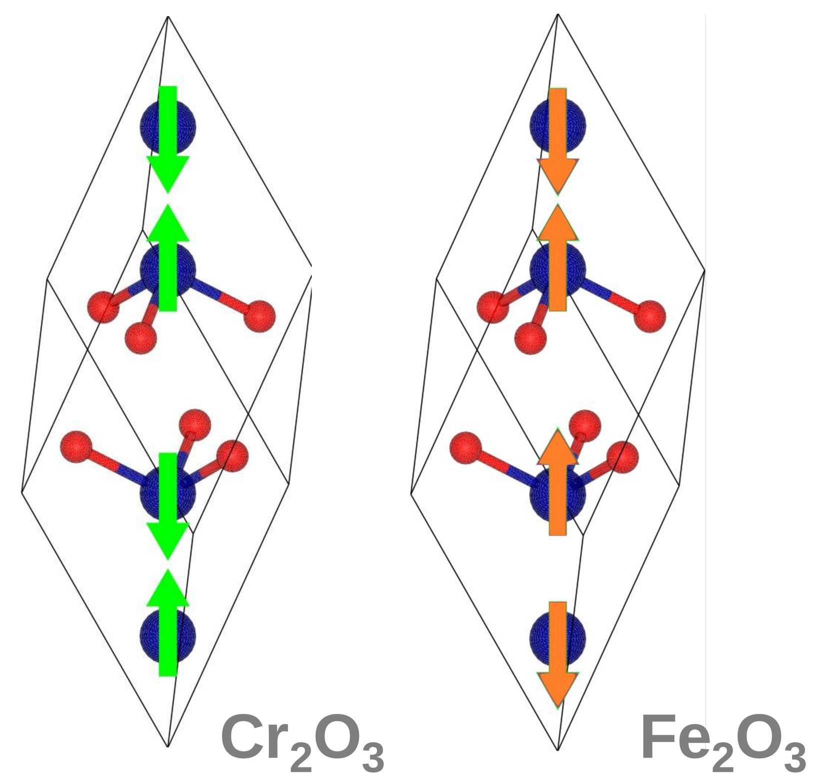
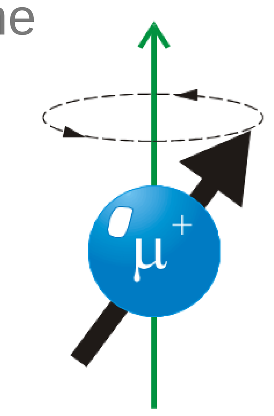
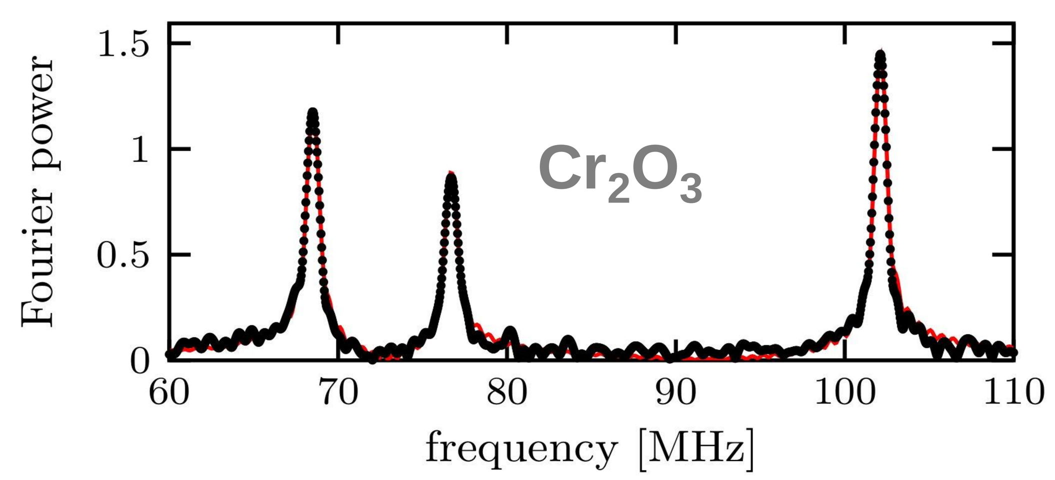
Hydrogen is one of the most ubi-quitous impurities in semiconductors and insulators, and has a significant impact on their electronic properties. Understanding the dopant charac-teristics of such defects is critical for a precise control of charge carriers, upon which modern electronic tech-nology is based. Since isolated hydrogen is extremely hard to study directly, most of the available infor-mation comes from the study of muonium, a charge-neutral bound state of a short-lived positive muon and an electron (Mu=[μ⁺e⁻]), which, having virtually an identical elec-tronic structure inside a material as interstitial hydrogen, serves as a light hydrogen analogue. In magnetic ma-terials, however, Mu is not observed, and charge-neutral muon states are generally not considered relevant.
We discovered charge-neutral complexes in antiferromagnetic Cr_2O_3 and Fe_2O_3, demonstrating that such states do exist in magnetic materials, and opening a route to study the electronic structure of hydrogen defects in magnetic oxides.



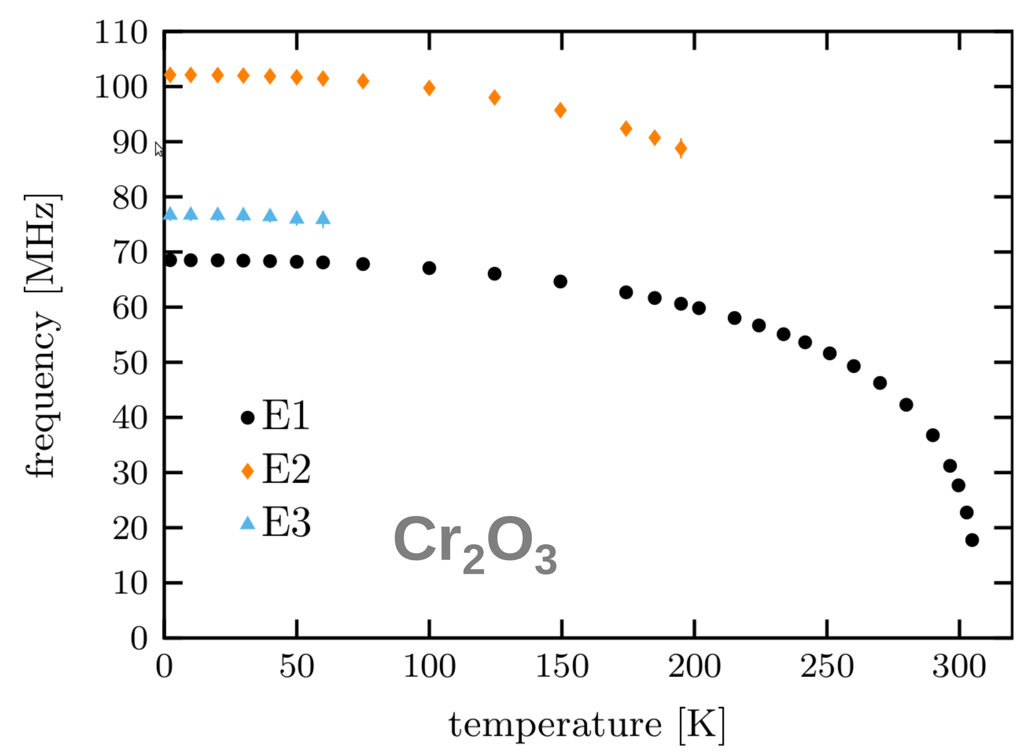
Muon Spin Rotation

- spin polarized, positively charged muons μ^+ are implanted into sample
- muon lifetime τ=2.2 μs: anisotropic emission of the decay positrons allows to observe muon spin polarization as function of time
- stopping sites in crystal lattice well defined, several different sites possible
- spin 1/2: spin precession in magnetic fields, which can be used as a direct measure of internal fields at muon stopping site

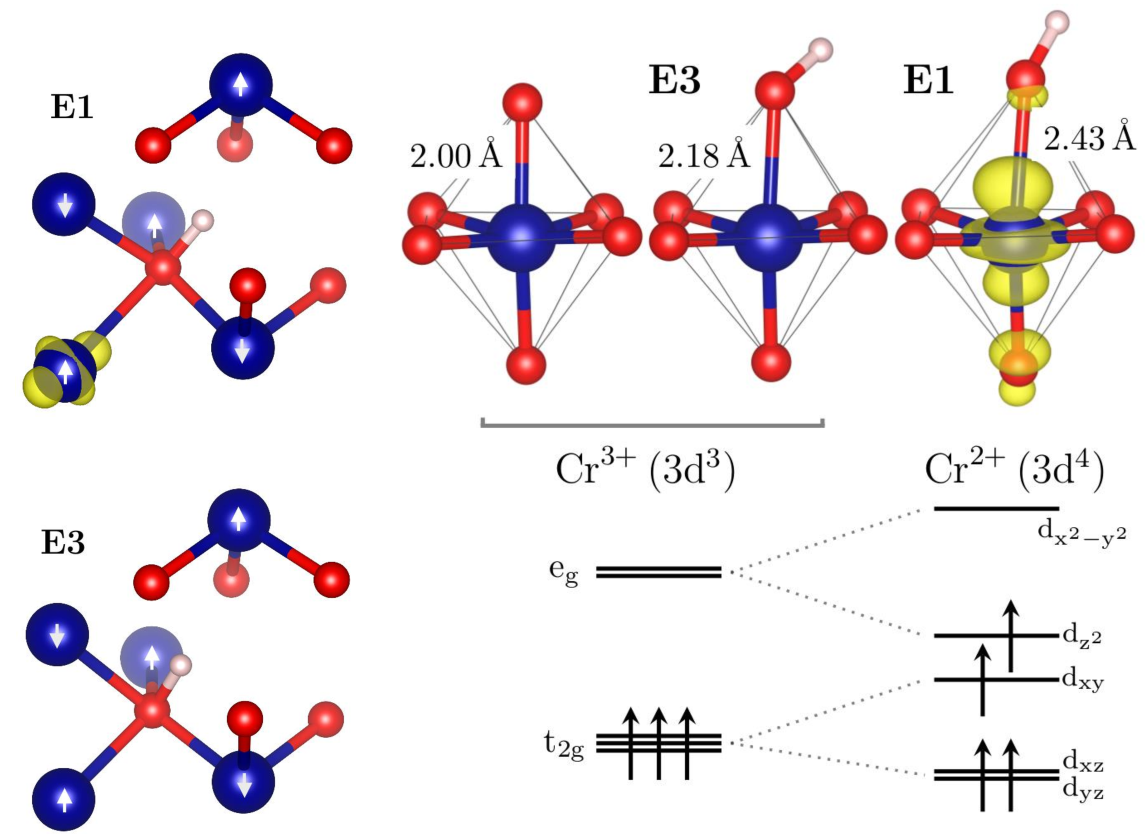




Fourier transform of a μSR spectrum in Cr_2O_3 at 5K



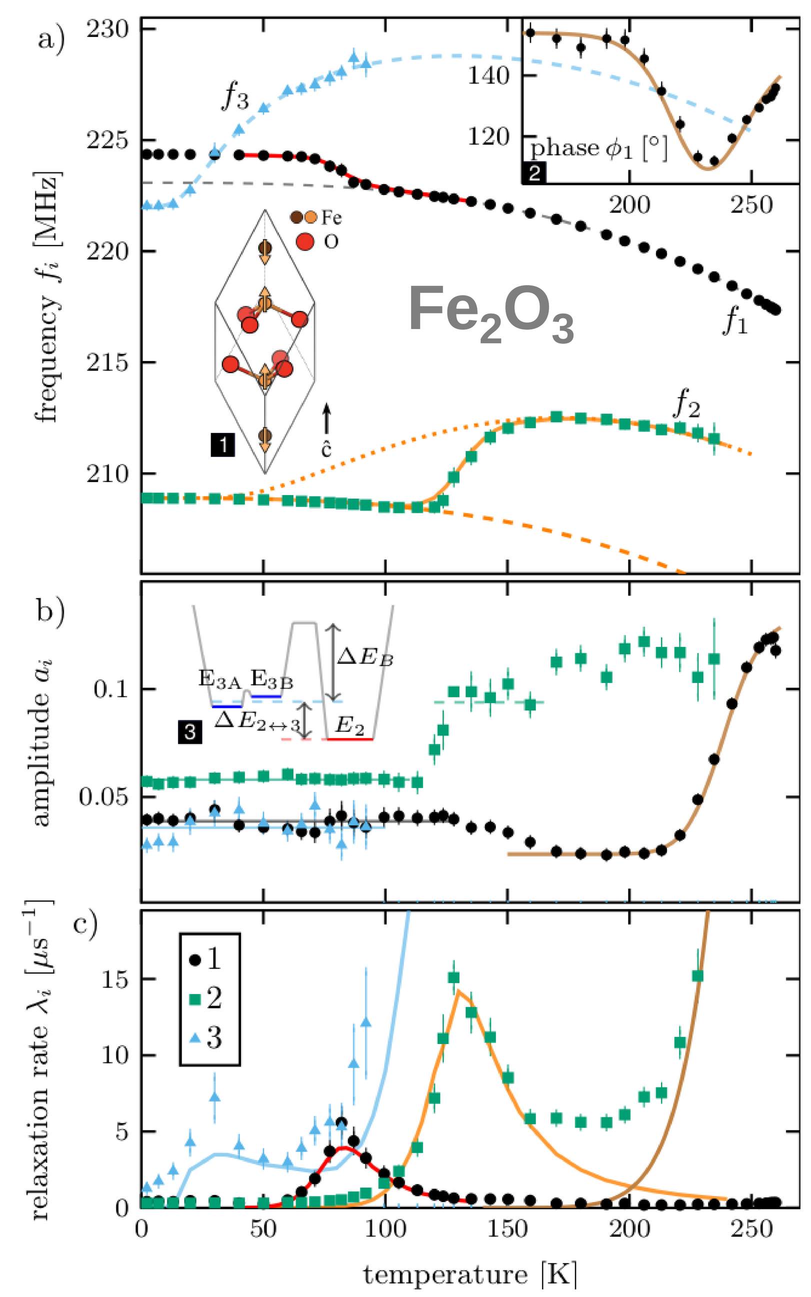
Zero field precession frequencies vs. temperature



Jahn-Teller stabilized muon-polaron complex: a neutral charge-state formed by the muon and an extra electron localized on the Cr, changing its valence state from Cr^{3+} to Cr^{2+}

μSR results

- Three distinct muon stopping sites with different temperature behavior are observed in both materials
- The temperature dependence reveals a rich dynamic behavior that is interpreted in the context of local muon hopping and thermally activated site transitions
- Coexistence of highly dynamic and site-stable muons at the same temperature is explained by the formation of a charge-neutral, Jahn-Teller-stabilized muon-polaron complex.
- The muon-polaron complex is "hidden", since bound electron is strongly coupled to 3d electrons of host ions; thus no signatures conventionally expected from muonium are displayed
- The complex ionizes in Fe2O3, indicating hydrogen acts as a shallow donor



Zero field precession frequencies and amplitudes in Fe_2O_3

Conclusion

- Observation of charge-neutral muon states in Cr_2O_3 [1] and Fe_2O_3 [2] strongly suggests that similar complexes exist in other insulating or semiconducting magnetic materials; this opens a route to study the dopant characteristics of hydrogen in magnetic transition metal oxides, a good understanding of which is crucial for a precise control of charge carriers in such materials.
- The formation of a charge-neutral complex significantly changes how the muon interacts with its host material, which has implications for the interpretation of μSR results, both past and future, in a broad range of materials

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[1] M. H. Dehn et al., Phys. Rev. X **10**, 011036 (2020)
[2] M. H. Dehn, J. K. Shenton et al., Phys. Rev. Lett. **126**, 037202 (2021)

Discovery, accelerated