Long-Range Magnetic Order in the Magnetodielectric Regime of Ce_2O_3

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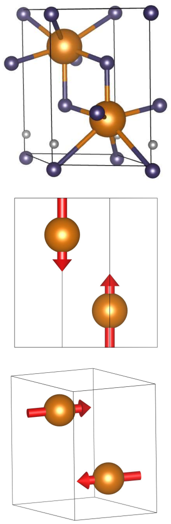
INTRODUCTION

The sesquioxide, Ce_2O_3, has been a material of intense interest in recent years due to reports of an anomalous giant magnetodielectric effect and the emergence of mixed crystal field-phonon (vibronic) excitations below a putative antiferromagnetic transition at T_N = 6.2K. The claim of long-range magnetic order in this material is based on heat capacity and temperature-dependent susceptibility measurements. Curiously, three previous neutron diffraction studies were unable to distinguish any magnetic Bragg peaks. To address this point, we undertook a combined study of polycrystalline Ce_2O_3 using neutron diffraction, triple-axis[1] and time-of-flight (TOF) inelastic neutron scattering[2] (INS) and muon spin rotation (µSR).[3]

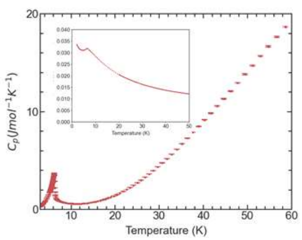


Crystal structure of Ce_2O_3 (top). Grey spheres reveal energetically favorable muon site. Proposed magnetic unit cell with easy-axis (center) and easy plane spins (bottom).

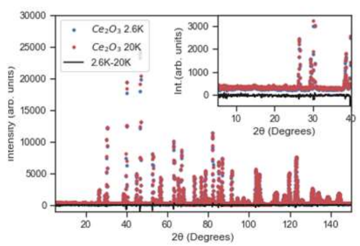
MEASUREMENTS

Heat capacity and magnetic susceptibility measurements imply the presence of a magnetic transition at 6.2K.

Heat capacity measurement shows lambda anomaly consistent with second-order transition. Dip in magnetic susceptibility measurement suggests onset of antiferromagnetic order (inset).



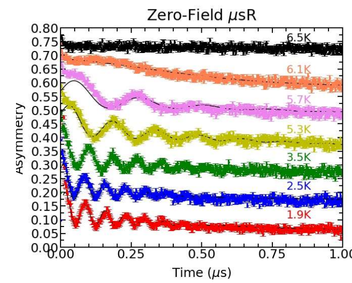
Neutron powder diffraction measurements were taken to investigate the purported magnetic order. No magnetic Bragg peaks appeared upon cooling below the transition.



Neutron powder diffraction taken at temperatures above and below transition. Black line shows difference curve.

RESULTS

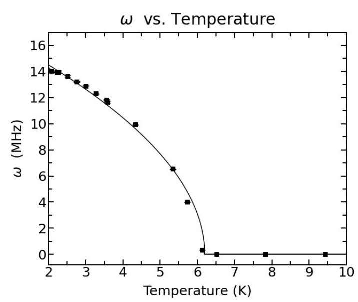
ZF-µSR measurements were carried out through the transition region to seek direct signatures of suspected magnetic order. This was confirmed by the emergence of clear oscillations below the transition temperature. Longitudinal field measurements indicate the order is static.



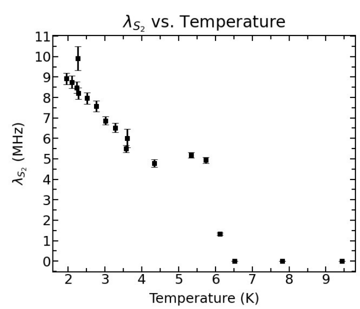
ZF-µSR asymmetry for powders of Ce2O3 at several temperatures around 6.2K Solid lines represent results of fits described below.

ZF data was fit to the depolarization function:

$$A_0P(t) = A_S(\tfrac{1}{3}e^{-\lambda_{s1}t} + \tfrac{2}{3}e^{-\lambda_{s2}t}\cos(\omega t + \phi)) + A_B(e^{-\lambda_{s1}t^{\beta}})$$



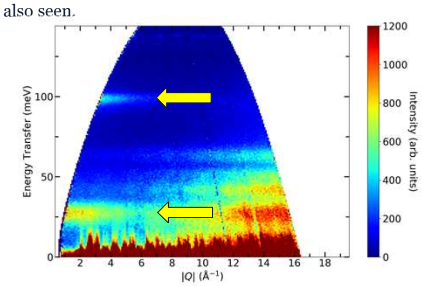
Oscillation frequency vs temperature. Solid line shows fit to the relation $|T-T_c|^{0.5}$.



Relaxation of oscillating component vs temperature.

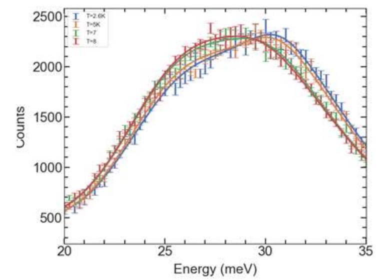
RESULTS

To determine the crystal field scheme and explore the emergence of vibronic modes, INS was performed using TOF and triple-axis instruments. Intensity at higher Q is associated with phonons, which agree with predictions for structural analogs. Magnetic excitations were observed at 27 and 99 meV, which are associated with CEF modes. At 27 meV we see scattering from both phonons and CEF modes which is consistent with reports from Raman scattering where an emergence of vibronic excitations was also seen.



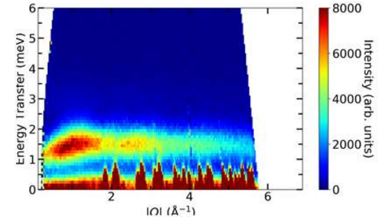
Measurements using E_i =160 meV neutrons reveal two crystal field modes. Yellow arrows indicate CEF modes.

While intrinsic peak broadening prevents us from observing vibronic modes directly, a we report a shift of spectral weight consistent with that seen in Raman studies.



Triple axis measurements in the region of expected vibronic modes.

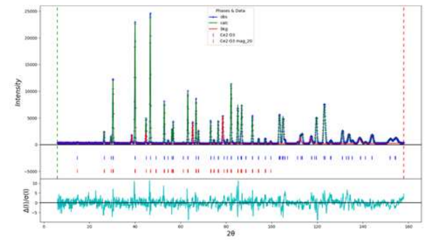
Moreover, spin waves appeared in TOF data upon cooling, providing further evidence for the existence of magnetic order below the transition.



Measurements using E_i =20 meV neutrons show emergence of spin waves below 6.2K.

DISCUSSION

Calculations were performed to determine the likely muon stopping site.[4] The lowest energy site was used to compare with data acquired from ZF-µSR. The local dipole field at this site was determined assuming the k=0 ordered state proposed in [5] with easy-axis spins. Based on the assumed magnetic order and the oscillation frequency from the ZF-µSR data, the size of the magnetic moment was calculated to be approximately 0.42μ_B. This small moment size would presumably explain the absence of magnetic Bragg peaks in the neutron data sets. Use of this moment size and ordered state to calculate a neutron powder diffraction pattern yields magnetic Bragg peaks that exist within the error bars of our data.



Neutron diffraction data. Solid line reveals calculated pattern from structural (blue ticks) and magnetic scattering (red ticks).

CONCLUSIONS

Presence of magnetic order in Ce_2O_3 has been observed using a combination of muon spin rotation and inelastic neutron scattering. Signatures of magnetic order emerged in the form of oscillations in the asymmetry of ZF-µSR measurements and spin waves in TOF inelastic neutron scattering. Muon site calculations assuming the magnetic order proposed in [5] and a Ce magnetic moment of 0.42 μ_B successfully reproduced the signal observed with µSR.

ACKNOWLEDGEMENTS

[1] Triple axis and diffraction data acquired at NCNR
[2] TOF data was collected at ISIS
[3] µSR data was acquired on M20 of CMMS at TRIUMF

[4] B. M. Huddart, A. Hernández-Melián, T. J. Hicken, M. Gomilšek, Z. Hawkhead, S. J. Clark, F. L. Pratt, and T. Lancaster, MuFinder: A program to determine and analyse muon stopping sites, arXiv:2110.07341
[5] Kolodiazhnyi et al. 10.1103/PhysRevB.98.054423

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