

Vibronic Coupling in a 4f Molecular Magnet Probed by Far-Infrared Magnetospectroscopy



J. G. C. Kragoskow,¹ J. Marbey,^{2,3} C. D. Buch,⁴ J. Nehrkorn,² M. Ozerov,² S. Piligkos,⁴ S. Hill,^{2,3} N. F. Chilton¹
1. University of Manchester, UK; 2. MagLab; 3. FSU Physics; 4. University of Copenhagen, Denmark

Funding Grants: G.S. Boebinger (NSF DMR-1644779); N. F. Chilton (Royal Society and European Research Council, grant # 851504); J. G. C. Kragoskow (EPSRC); S. Hill (DOE DE-SC0020260); S. Piligkos (VILLUM FONDEN #13376).

Vibronic coupling, the interaction between molecular vibrations and electronic states, is a fundamental effect that profoundly influences a wide range of physical processes. In molecular magnetic materials, vibronic coupling causes relaxation, which equates to loss of magnetic memory in single-molecule magnets and loss of phase coherence in qubits.

The study of vibronic coupling is challenging, and most experimental evidence is indirect. In this investigation, far-infrared magnetospectroscopy (FIRMS) is used to directly probe electronic transitions coupled to vibrational excitations in an ytterbium molecule (inset of Figure) that has attracted interest as a potential spin qubit. High magnetic fields (Figure) enable a deconvolution of vibronic transitions - transitions that shift in energy due to the Zeeman interaction - from a forest of much stronger electronic transitions that do not shift with field.

MagLab users found that vibronic coupling is strongest for vibrational modes that simultaneously distort the first coordination sphere (the atoms that coordinate directly to the ytterbium ion) and break the C_3 symmetry of the molecule. With this knowledge, vibrational modes could be identified and engineered to shift their energy towards or away from particular electronic states to alter their impact. Hence, these findings provide new insights towards developing general guidelines for the control of vibronic coupling in molecules.

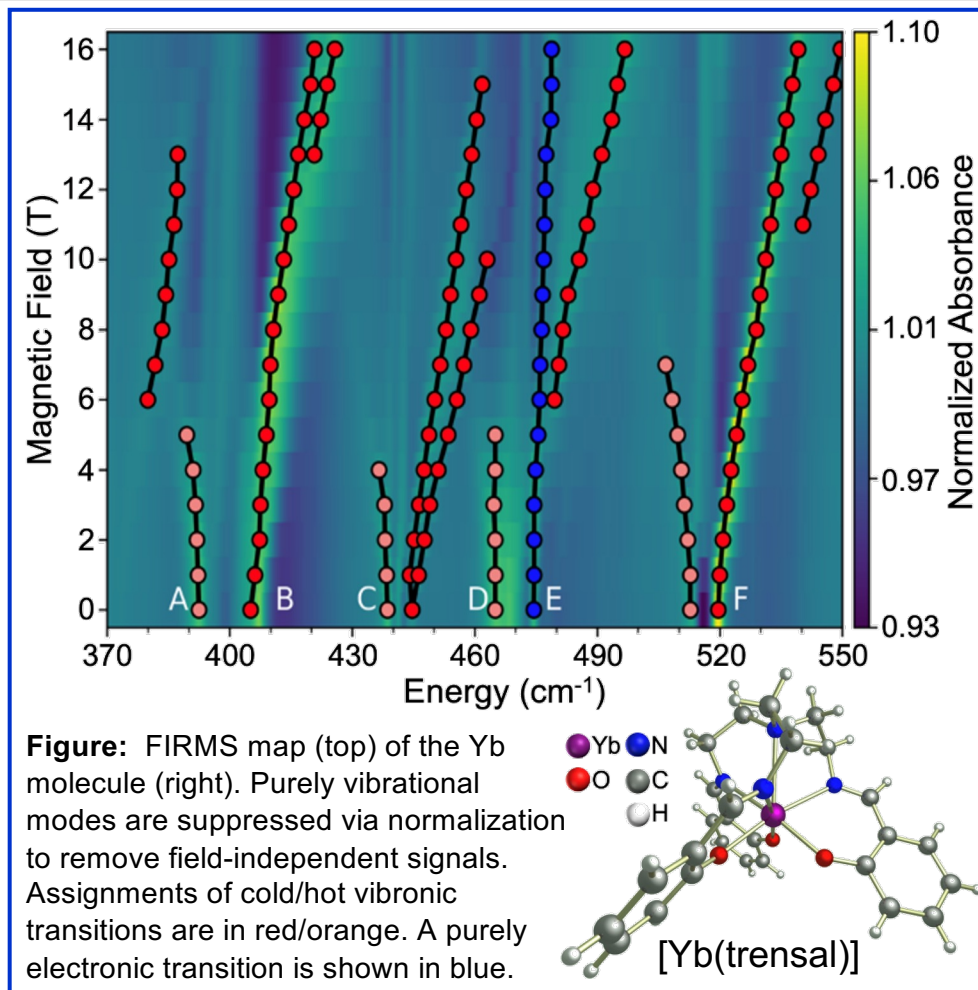


Figure: FIRMS map (top) of the Yb molecule (right). Purely vibrational modes are suppressed via normalization to remove field-independent signals. Assignments of cold/hot vibronic transitions are in red/orange. A purely electronic transition is shown in blue.

Facilities and instrumentation used: Joint EMR/DC-Field Facility Operation (Infrared / Terahertz Magneto Optics in SCM3)

Citation: Kragoskow, J.G.C.; Marbey, J.; Buch, C.D.; Nehrkorn, J.; Ozerov, M.; Piligkos, S.; Hill, S.; Chilton, N.F., *Analysis of vibronic coupling in a 4f molecular magnet with FIRMS*, **Nature Communications** **13**, 825 (2022) doi.org/10.1038/s41467-022-28352-2 Links to Data [Set 1](#) and [Set 2](#)