

Optical study of vibronic coupling in the quantum spin liquid candidate $\text{Tb}_2\text{Ti}_2\text{O}_7$

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In the study of geometrically frustrated magnetism, the precise nature of the ground state in $\text{Tb}_2\text{Ti}_2\text{O}_7$ has remained a long-standing conundrum. In this pyrochlore material, no conventional spin-ice or long-range magnetic order is stabilized, even at very low temperatures. Quantum fluctuations are suspected of being at the origin of an exotic phase, yet so far they have lacked conclusive evidence. Using high-resolution synchrotron-based terahertz spectroscopy, we have probed the lowest energy excitations of $\text{Tb}_2\text{Ti}_2\text{O}_7$. It is revealed that a double hybridization of crystal-field-phonon modes is present across a broad temperature range. This so called vibronic process affects the electronic ground state that can no longer be described solely by electronic wave functions. Rather, a collective state prevails, built on the ground and first excited crystal-field states mixed with two different phonon modes. This provides a crucial path for quantum spin-flip fluctuations to inhibit the stabilization of conventional magnetic states. The study is further complimented by recent ultrafast terahertz-pump/optical-probe measurements on the ELBE free electron laser that confirm the presence of the vibronic coupling.

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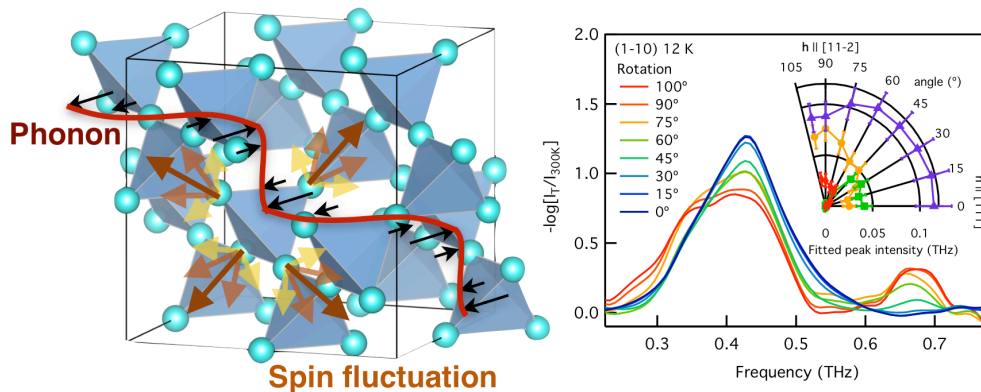


Figure 1 : (left) Tb^{3+} Pyrochlore network and graphical description of the vibronic process that involves the hybridization between a phonon and a crystal field excitation. (Right) Angular dependence of the THz spectra.