Nearly-Resonant Crystalline-Phononic Coupling in Quantum Spin Liquid Candidate CsYbSe$_2$

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Abstract: We study crystal field-phonon interactions in quantum spin liquid candidates CsYbSe$_2$ with variable temperature, polarization, and magnetic-field dependent Raman microscopy. Complex interplay between phonon modes and CEF modes is observed in real space. © 2022 The Author(s)

The quantum spin liquid (QSL) [1, 2] is a theoretically proposed phase characterized by many-body entanglement. The realization of a QSL phase may ultimately enable fault-tolerant quantum information processing, though conclusive evidence of many-body entanglement in QSLs remains a challenge. Among possible candidate materials, Yb-delafossites [3] have been under intensive study recently due to their ideal triangular lattice and less defect-prone nature. 14 members of this family including NaYbSe$_2$ [4-7], KYbSe$_2$ [8] and CsYbSe$_2$ [9, 10] have been identified and none has shown long-range order at zero magnetic field and low temperatures.

![Spatial dependent Raman spectra for CsYbSe$_2$ at $T = 3.3$ K comprising 45 by 45 pixels over 96 by 88 $\mu$m. (a) 2.025 raw spectra. Inset: non-negative matrix factorization (NMF) basis components (vectors). (b)-(e) spatial dependence of integrated counts over selected peaks. (e) spatial dependence of total counts. (f)-(g) spatial map of NMF weights. Adapted from Ref. [11].](image)

Here, we study temperature-, polarization-, and magnetic-field-resolved Raman microscopy of CsYbSe$_2$. We identify all the primary phonon modes and crystal electric field (CEF) modes in CsYbSe$_2$, and we demonstrate
polarization dependence consistent with the P6/mmmc (No. 194) assignment [11]. The number of primary CEF levels, which come from the J = 7/2 manifold of Yb$^{3+}$ ions, is consistent to the environment of the Yb$^{3+}$ ions are in.

Figure 1 shows the spatial dependence of the Raman spectra over a 96 by 88 μm area. While raw integration is vulnerable to nearby modes, NMF decomposition of the Raman map illustrated spatial anticorrelations between modes: for example, in the inset of Fig. 1(a), the NMF basis component 1 (red) is characterized by a wide peak around 233.9 and 255.4 cm$^{-1}$, which are identified as CEF2+E$^{1}_{2g}$, and 2E$^{2}_{2g}$, respectively, while the basis component 2 (blue) is characterized by CEF1-3 modes. Surprisingly, as seen in (f) and (g), the spatial weight for basis components 1 and 2 are anticorrelated.

Additional polarization and magnetic-field dependent Raman microscopies of CsYbSe$_2$ performed at 4 K and in fields of -6 to 6 T illustrated a strong field dependence for circularly polarized measurements. The ability to control CEF-phonon interactions in these candidate quantum spin liquids with mesoscale magneto-optical probes may ultimately provide a pathway to optical control of QSL-based quantum devices.

This research was sponsored by the U. S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Sciences and Engineering Division. The first principles phonon calculations and Raman microscopy were performed at the Center for Nanophase Materials Sciences, which is a U.S. Department of Energy Office of Science User Facility. L.L. acknowledges computational resources of the Compute and Data Environment for Science (CADES) at the Oak Ridge National Laboratory, which is supported by the Office of Science of the U.S. Department of Energy under Contract No. DE-AC05-00OR22725. Postdoctoral research support was provided by the Intelligence Community Postdoctoral Research Fellowship Program at the Oak Ridge National Laboratory, administered by Oak Ridge Institute for Science and Education through an interagency agreement between the U.S. Department of Energy and the Office of the Director of National Intelligence.

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