Spectroscopic Analysis of Quantum Spin Liquids and Nanophotonic Cavities

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Introduction

The spectroscopy of quantum materials and plasmonic heterostructures produces multidimensional and hyperspectral datasets containing information about the magnetic, electronic, and optical properties of a sample. In this project, we explore datasets acquired from two classes of materials: quantum spin liquids (QSL) and nanophotonic cavities.

QSL's are a type of quantum material that exhibits long-range disorder due to the frustration of their magnetic moments. Their exotic properties, such as long-range entanglement and fractional spin excitations, have application potential for quantum communication and topological quantum computation.[1]

Nanophotonic cavities, such as the plasmonic nanopatch antenna presented here, have been studied widely using far-field optical spectroscopy. Here, we study the near-field optical response to gain a better understanding of the plasmon-emitter interaction for future applications in functional nanophotonic devices.

Experimental Methods

Raman Spectroscopy: In our study of α-$RuCl_3$, we apply magneto-Raman spectroscopy over temperatures from T = 670 mK to T = 4 K and 532 nm excitation wavelength, since it can detect microscopic phonon-magnon and phonon-spinon coupling, which can be used to identify potential QSL states.

Cathodoluminescence Microscopy: CL Microscopy directly probes the radiative local density of optical states. We excite our plasmonic heterostructures with a 30 keV electron beam and collect the resulting emission at small and large fields of view to obtain interferograms and maps of localized plasmon modes

Scanning Electron Microscopy: A Scanning Electron Microscopy (SEM) image is acquired as a byproduct of sweeping a converged electron beam over the sample during CL microscopy. It can be used to study correlations between morphology and dynamics in a material and be applied for drift correction.

Data Analysis Methods

Nonnegative Matrix Factorization: Nonnegative matrix factorization (NMF) is a spectral unmixing method that extracts sparse and meaningful features from a set of nonnegative data vectors. In the context of spectroscopy, NMF can decompose a spectral data matrix Y as,

$$Y \approx WH \qquad (1)$$

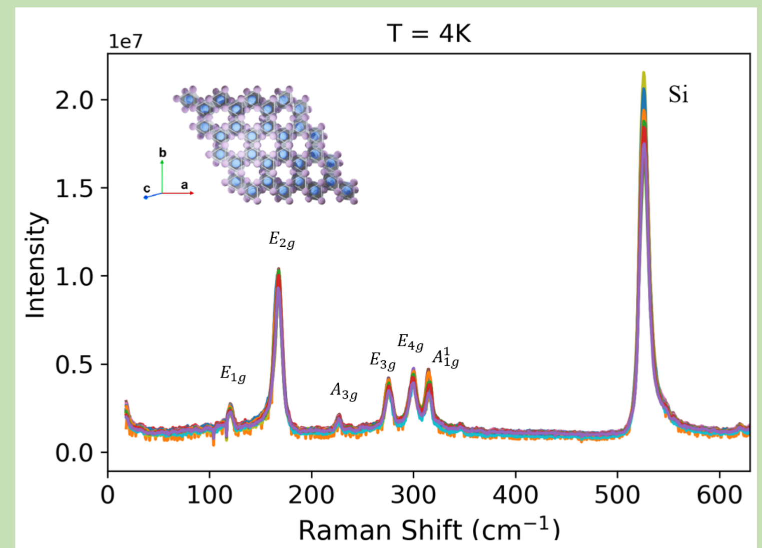
Where W contains the spectral endmembers and H contains the relative abundance of each endmember.

Python Packages: HoloViews, an open-source Python library, was used for data visualization and was used in conjunction with the Panels package to develop a user interface for processing and analyzing CL data. Sci-kit learn, a Python machine learning library, was used to perform NMF decomposition.

Results

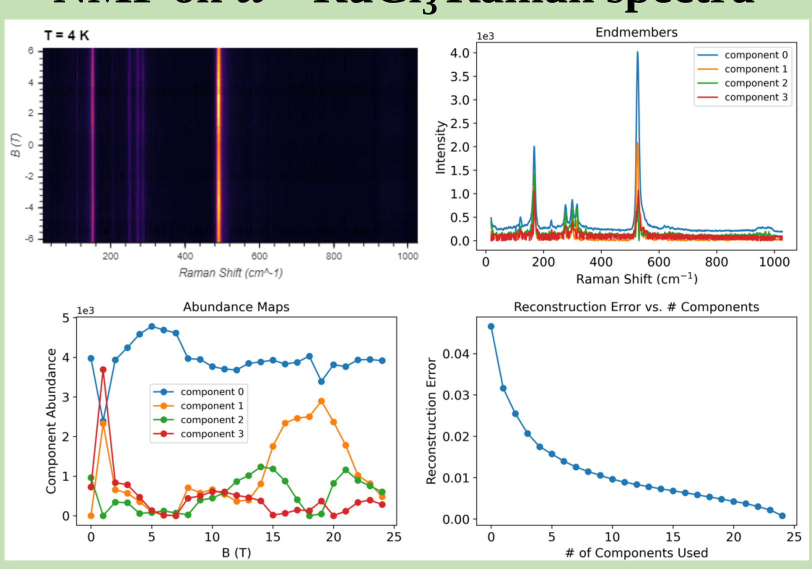
Spectral Unmixing of Multidimensional α-$RuCl_3$ Raman Spectra

$\alpha-RuCl_3$ Raman Modes



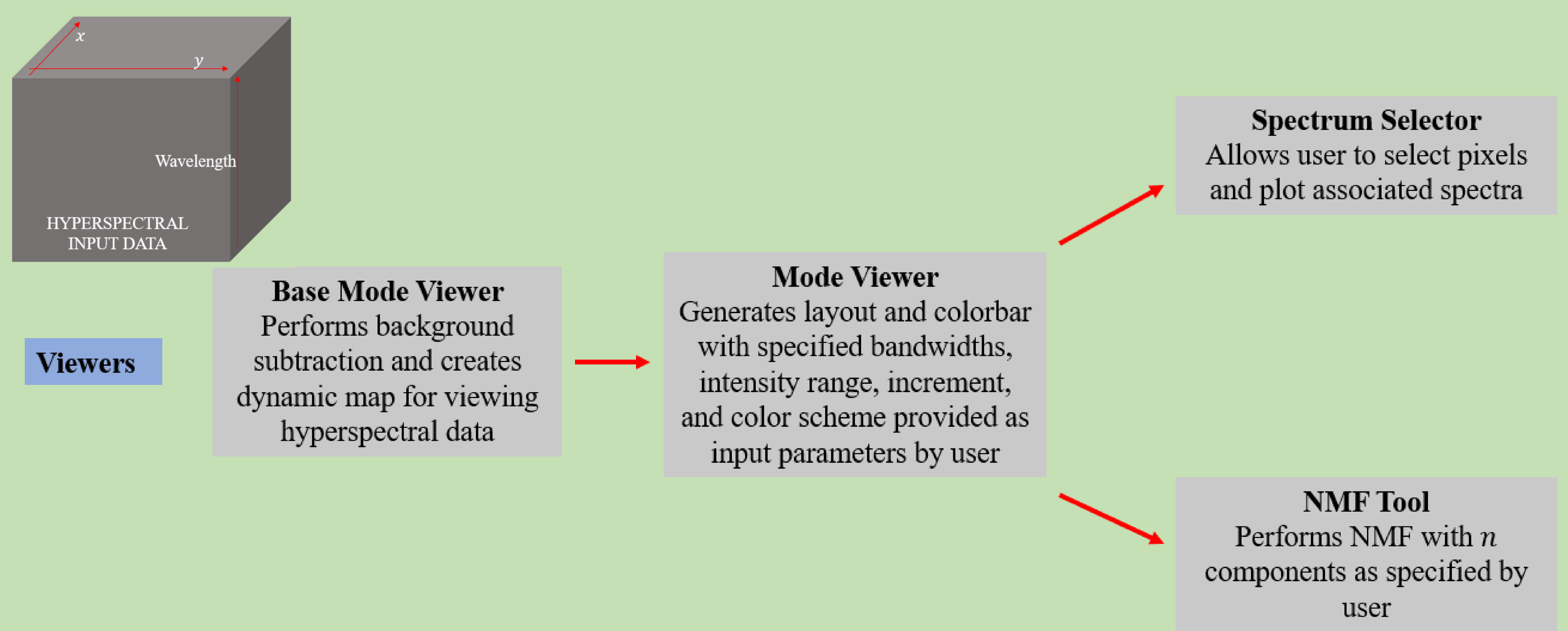
Due to the honeycomb lattice structure of the material α-$RuCl_3$, the Kitaev model predicts that its zigzag ground state may harbor a QSL.[1] The temperature-dependent Raman datasets are processed by removing the baseline via an asymmetric least-squares algorithm. Baseline analysis (not shown) is performed by frequentist fitting.

NMF on $\alpha-RuCl_3$ Raman spectra

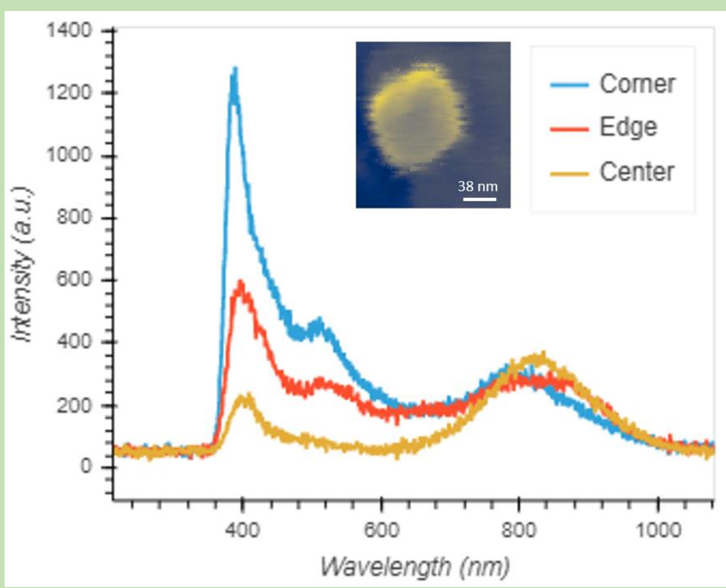


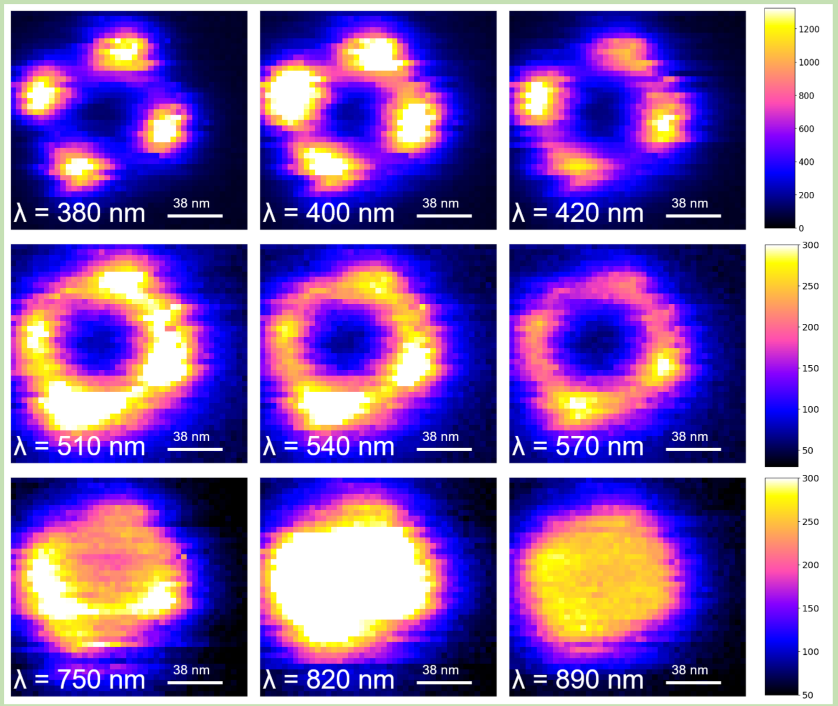
We decompose the Raman spectra by NMF as shown in the example on the left with $n = 4$ components. To address the difficulty in determining the number of NMF components to use *a priori*, we study the NMF reconstruction error, which shows a leveling-off in error decrease with increasing number of components, starting at about nine components. Ultimately, we chose $n = 4$ components to avoid noise-dominated endmembers.

Visualization and Analysis of Hyperspectral Cathodoluminescence Datasets on Ag Nanopatch Antenna



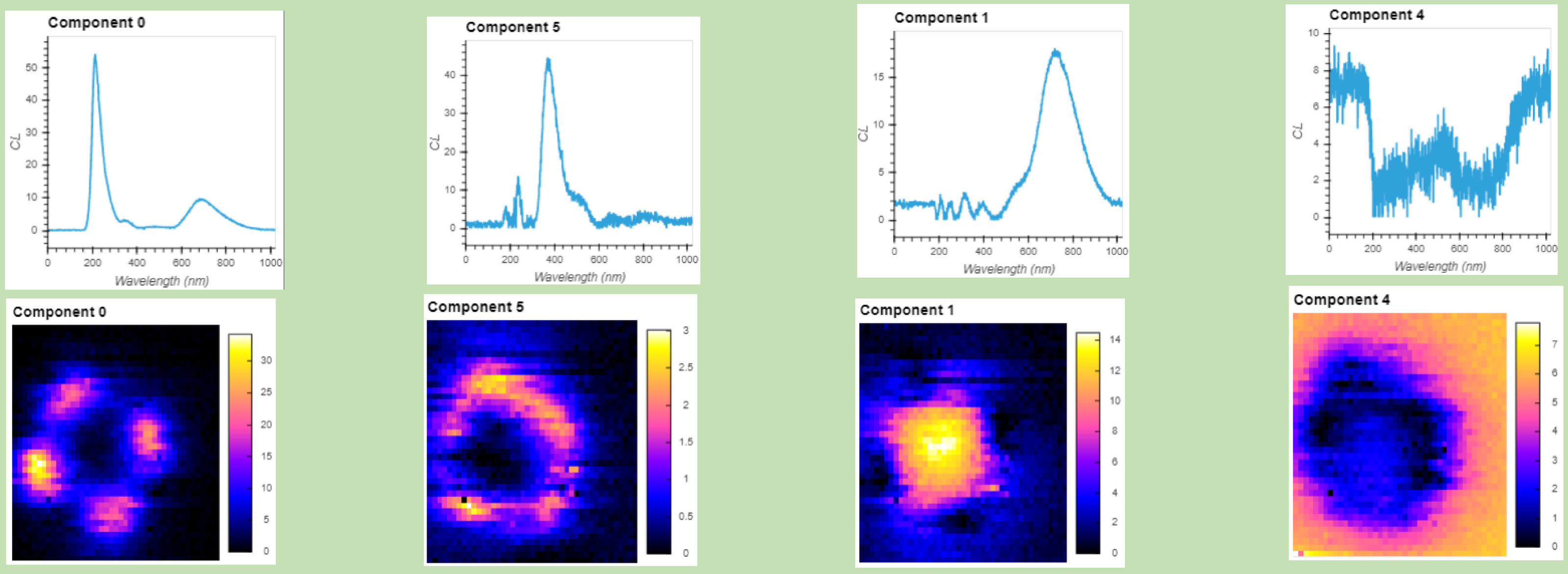
Framework: We developed a Python panel that allows the user to dynamically visualize plasmon modes in our CL dataset on our Ag Nanopatch Antenna by interactively adjusting filtering parameters. The tool also allows the user to select spectra from pixels and perform a cursory NMF.



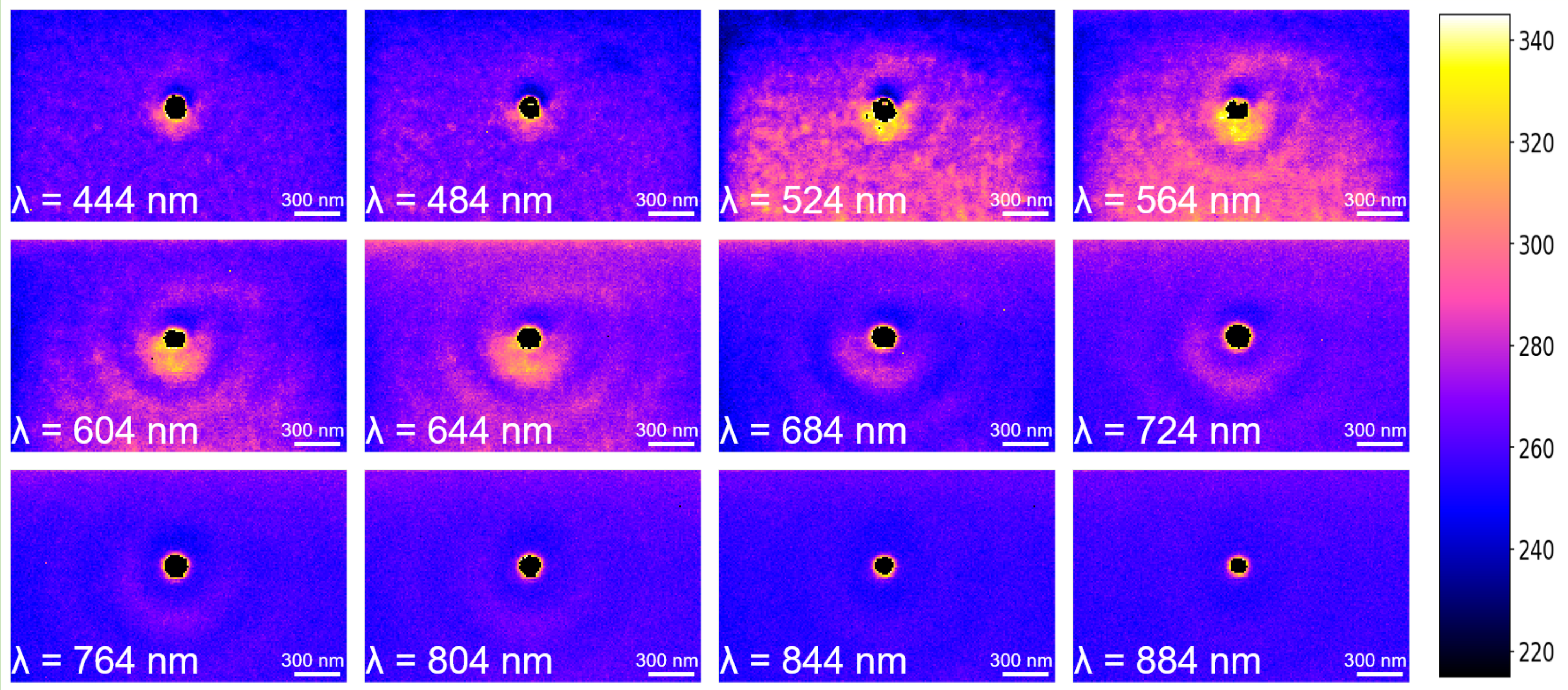


Localized Plasmon Modes: Three distinct plasmonic modes can be observed at high magnification: a corner mode, an edge mode, and a bulk mode, which are given by the peaks in the spectrum on the left. The corresponding CL maps are shown in the plot layout on the right.

Corner | Edge | Bulk | Other



NMF: NMF endmembers with corresponding abundance maps depict corner modes, edge modes, and bulk modes that can be compared with the experimentally acquired spectra from the corner, edge, and center of the nanocube. Shown above are examples of components from NMF with $n = 20$ components. Many of the other components are noise dominated or are attributed to nonlinear processes.



Interferograms: Impinging electrons from CL generate broadband transition radiation in the substrate, as well as propagating surface plasmon polaritons (SPP) that scatter from the Ag nanocube.[2,3] A clear pattern resulting from the interference between the SPPs and transition radiation is observed in the interferograms above.

References
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[2] Kuttge, M., et al. (2009). "Local density of states, spectrum, and far-field interference of surface plasmon polaritons probed by cathodoluminescence." Physical Review B **79**(11).
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