Unraveling the Correlations between Raman and Photoluminescence in Monolayer MoS_2 through Machine-Learning Models

Masao Horiba Awards

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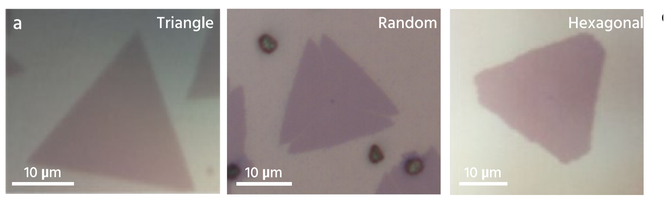
Rationale

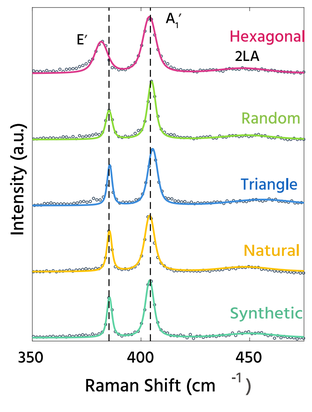
2D semiconductors with intense and tunable PL have opened up new opportunities for optoelectronics such as LED and single-photon emitters. Raman spectroscopy is a fast and non-destructive technique capable of probing material crystallinities and perturbations such as doping and strain. However, due to its highly nonlinear nature, a comprehensive understanding of the correlations between PL and Raman spectra in MoS_2 remains elusive.

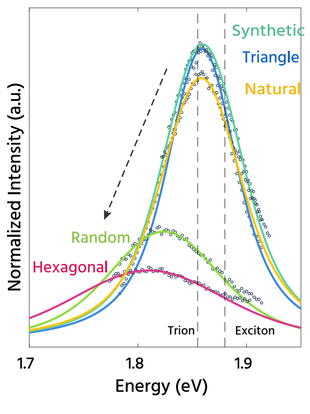
Summary

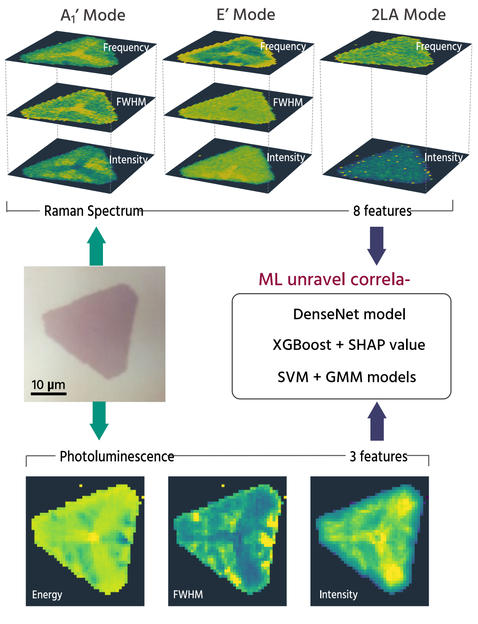
This work systematically explores the connections between PL signatures and Raman modes. First, we deploy a DenseNet to predict PL maps by spatial Raman maps. Moreover, we apply a gradient-boosted trees model (XGBoost) with Shapley additive explanation (SHAP) to bridge the impact of individual Raman features in PL features. Last, we adopt a support vector machine (SVM) to project PL features on Raman frequencies.

Sample and Data Preparations of Raman and PL Spectra



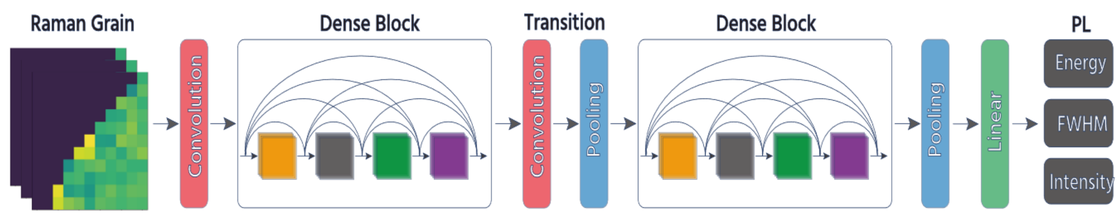


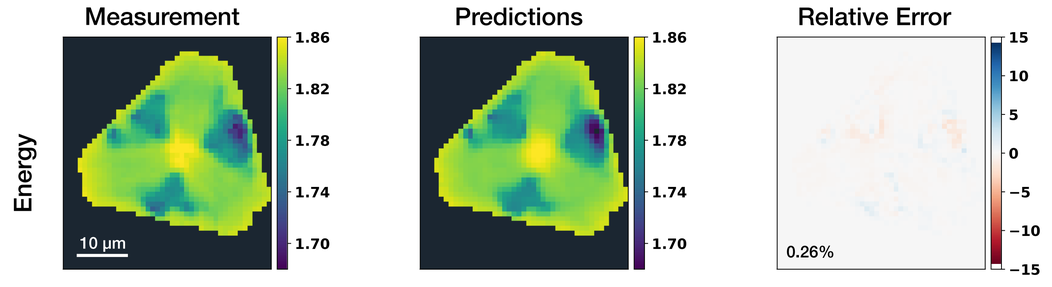




- MoS_2 crystals are prepared by exfoliation and chemical vapor deposition methods
- Oxygen levels increase from triangle, random, to hexagonal shapes
- Triangle CVD-grown and synthetic exfoliated MoS_2 crystals exhibit better PL performance than others
- PL and Raman spectra can be described by Voigt functions with three parameters – position, intensity, FWHM – for dimensional reductions

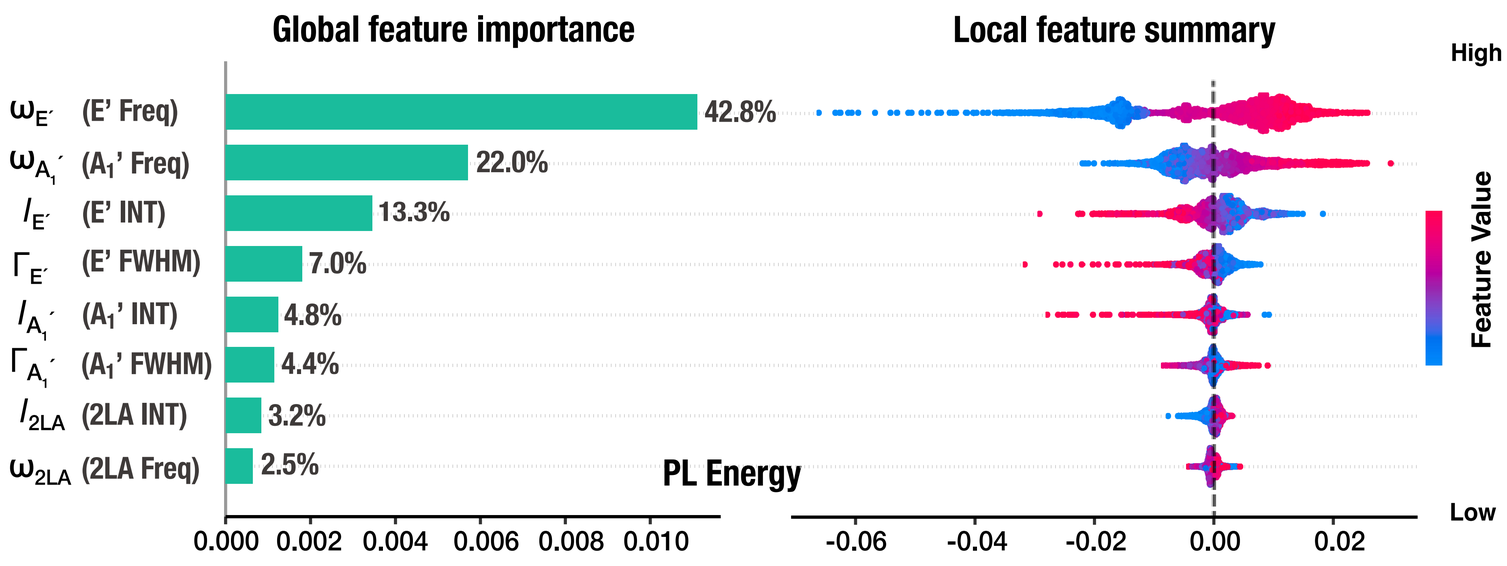
DenseNet Performs High Accuracy PL Predictions





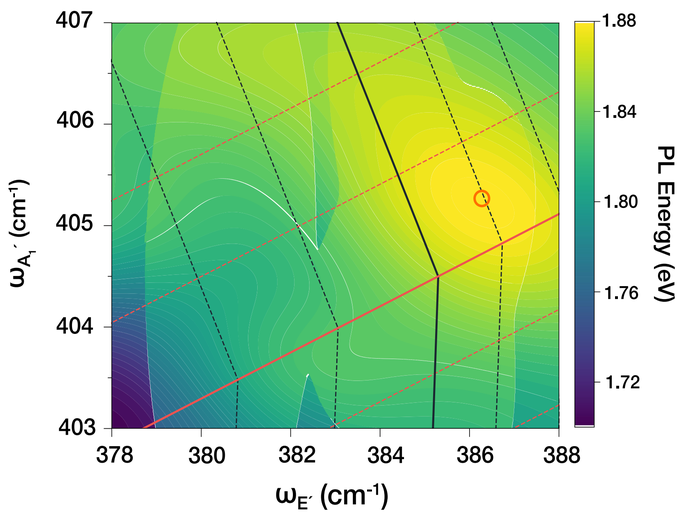
- Spectra maps are high-dimensional images
- 5-by-5 patch size shows the highest accuracy than other patched sizes
- PL energy and FWHM show higher accuracy
- PL intensity prediction shows unideal measurement conditions and fitting process

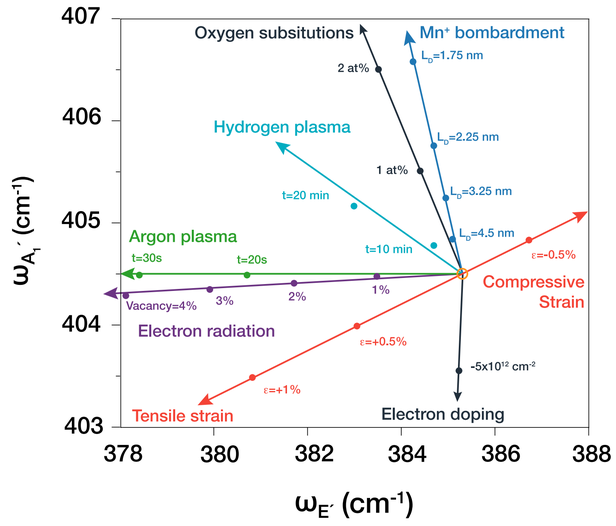
XGBoost with SHAP Values Explains the Correlations



- **Intrinsic Effects**
  - width ↑ , lifetime ↓ , crystallinity ↓
- **Doping Effects**
  - A1′ Freq. ↓ , Electron doping ↑ , Fermi level ↑
- **Strain Effects**
  - E′ Freq. ↓ , Tensile strain ↑ , bandgap transition (indirect ↑)
  - 2LA intensity ↓ , Tensile strain ↑

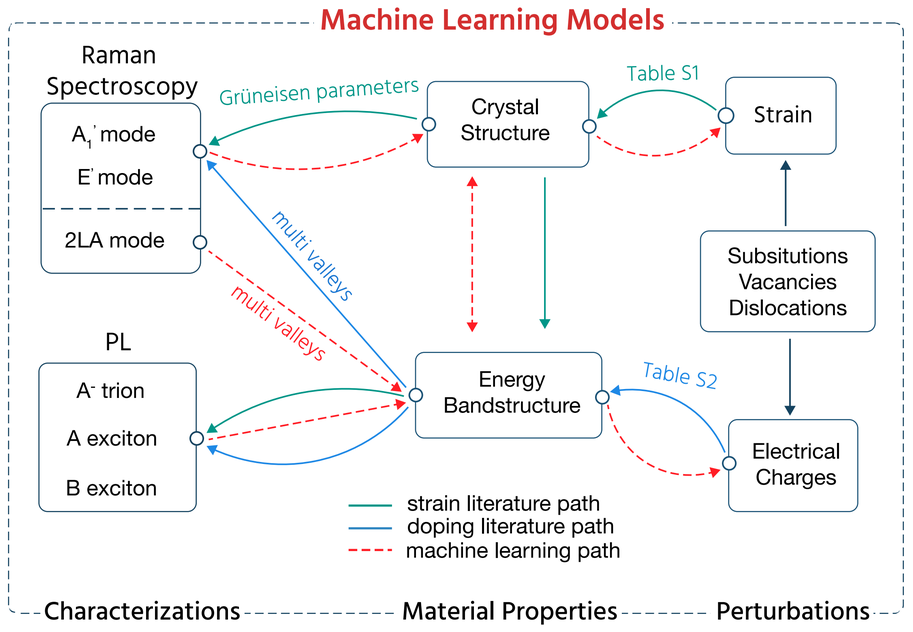
Visualize PL-Raman Correlations on Frequency Plot





- SHAP explainer indicates E′ and A_1′ freq. are the dominant factors
- Support vector machine (SVM) predicts PL behaviors on the frequency plot
- Frequencies-plot shows external-perturbation and defect-structure vectors

Overviews



In the schematic diagram (left panel), the knowledge path of the machine learning models (the red line), starts from characterization results via existing material knowledge to external perturbations and defective structures. This approach allows us to adopt the previous studies investigating Raman and PL spectra changes by controlling a single external perturbation, i.e., strain (the green line) or doping (the blue line), to form comprehensive knowledge of MoS_2 monolayers.

Prospective

The proposed methodology establishes an analysis approach to comprehensively interpret experimental observations to explore novel physics, which is suitable for Raman spectra and PL on 2D materials and for many other types of spectroscopies and condensed matter. In the ultimate goal, this methodology can be integrated into an autonomous platform for expediting advanced material manufacturers in the semiconductor industry.

Lu A-Y, Martins LGP, Shen P-C, Chen Z, Park J-H, Xue M, et al. Unraveling the correlation between Raman and photoluminescence in monolayer MoS2 through machine-learning models. Adv Mater. 2022;34: e2202911.