Supplemental Material: Observation of correlated plasmons in low-valence nickelates

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CONTENTS

S1. RESONANT BEHAVIORS OF PLASMONS

The overdoped low-valence nickelates $R_4Ni_3O_8$ exhibit a pre-peak feature in the O K-edge XAS [1], indicative of an appreciable amount of ligand holes and nonzero Ni-O hybridization. Figure S1 depicts the RIXS energy map measured on $Pr_4Ni_3O_8$ single crystals, which shows multiple Raman-like features resonant at the pre-peak regime, indicating various excitations. As revealed in our prior studies, the high-energy strong features correspond to charge-transfer excitations, and the lower-energy weak features originate from dd excitations through Ni-O hybridization. Plasmons are observed with lower energy loss ($\sim$0.2 eV), which is absent in the insulating phase of $La_4Ni_3O_8$ [2]. Its resonant behavior highlights the localized characters.

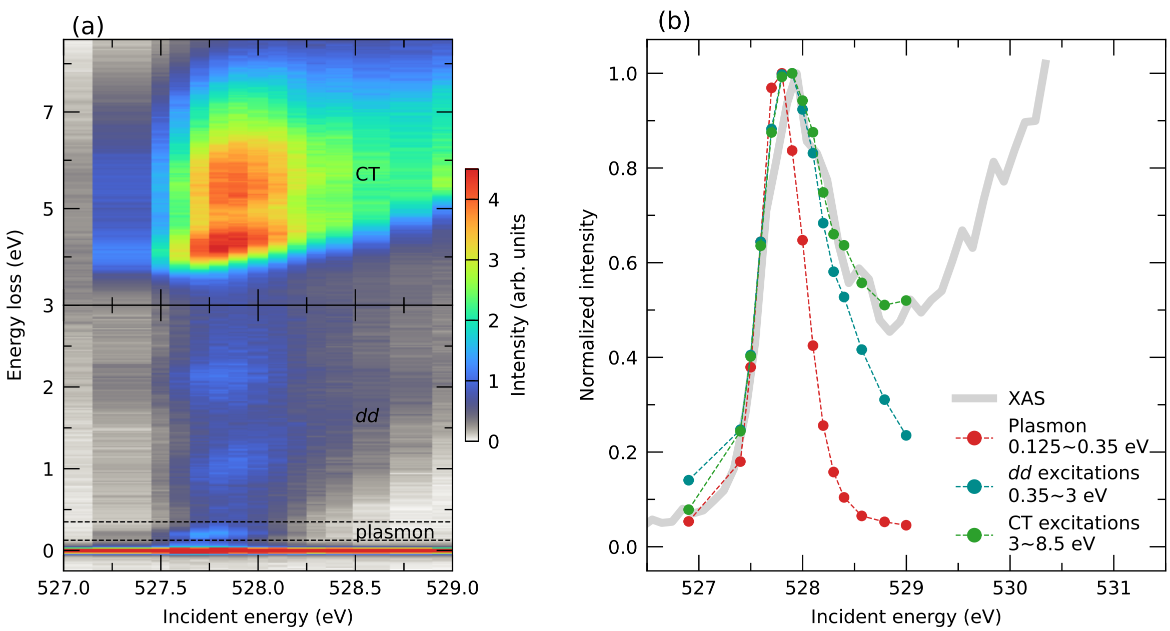

FIG. S1. Energy dependent resonant inelastic x-ray scattering (RIXS) spectra. (a), RIXS energy maps across the pre-edge regime of the O K edge collected with θ=22.6 ° and σ polarization. Various excitations are identified, including the charge-transfer (CT) excitations, orbital dd excitations, and plasmons. Additionally, weak phonons are observed overlapping with the quasi-elastic peak. The XAS data were collected in partial fluorescence yield (PFY) mode. (b), Incident energy dependence of the RIXS spectral weights integrated in the indicated energy ranges, representing different excitations.

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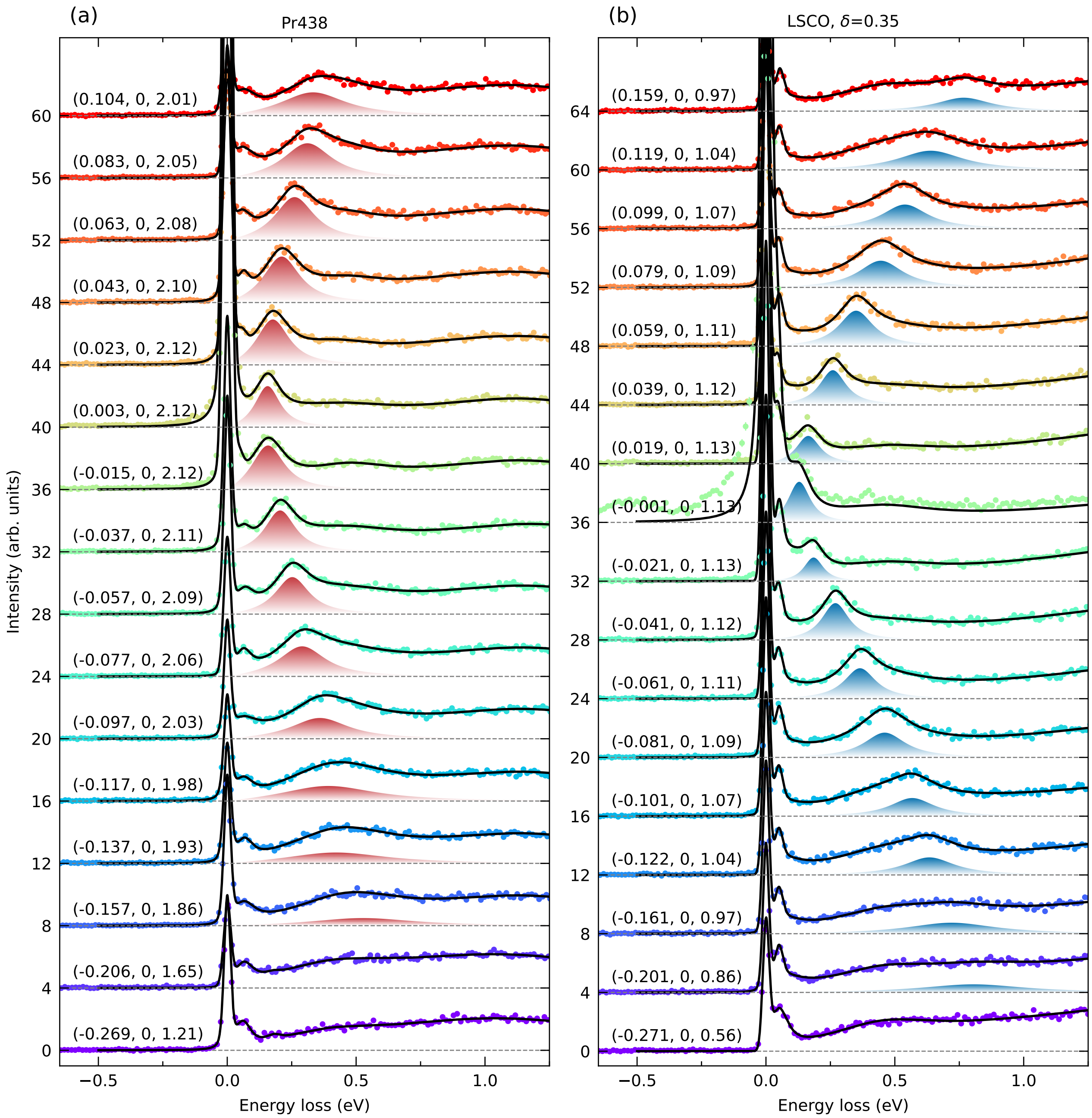

FIG. S2. Background subtracted O K-edge RIXS spectra for the in-plane plasmon dispersions. All the data for $Pr_4Ni_3O_8$ (Pr438) and $La_{2-x}Sr_xCuO_4$ (LSCO) were measured at 40 K. The dots present the data collected at the indicated $\mathbf{Q}$ positions, and the solid lines are fitting results. The shaded areas highlight the plasmon contributions.

S2. FITTING OF THE RIXS SPECTRA

Figure S2 and Fig. S3 present the RIXS spectra for plasmon dispersions along the in-plane and out-of-plane directions, respectively. A constant background has been subtracted for all the spectra using the energy-gain side. For $Pr_4Ni_3O_8$, each spectrum was fit with five components, including a pseudo-Voigt profile, representing the quasi-elastic peak, and four damped harmonic oscillators convoluted with resolution functions determined by the quasi-elastic peak to account for the phonons at $\sim$0.08 eV, dispersive plasmons indicated by the shaded areas, and two high-energy dd excitation modes. For $La_{2-x}Sr_xCuO_4$, we consider two phonon modes, one dispersionless bimagnon branch, and a tail from higher-energy charge excitations.

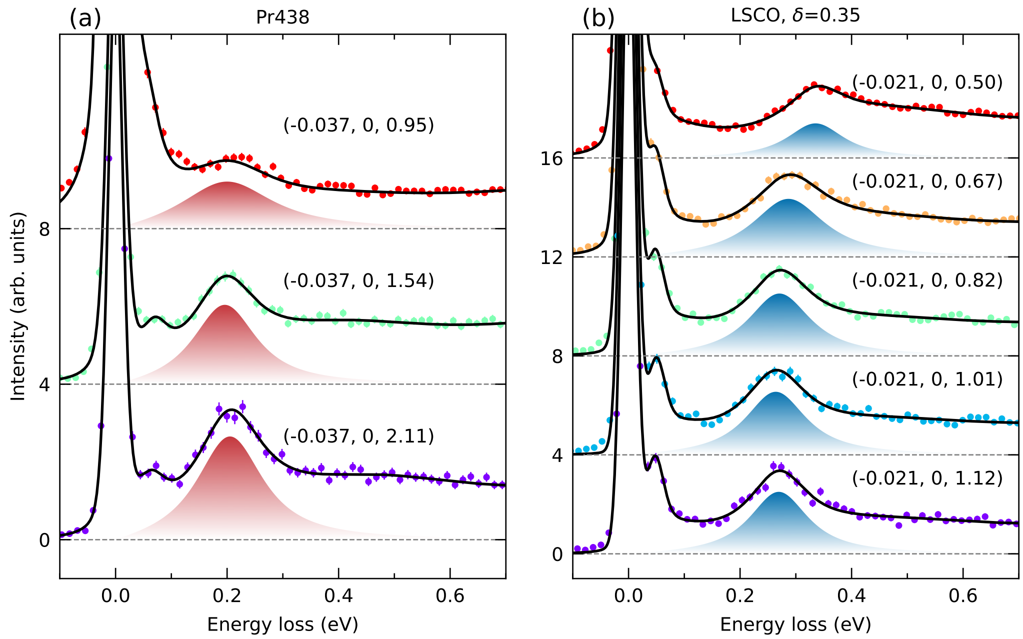

FIG. S3. RIXS spectra along the the out-of-plane direction.

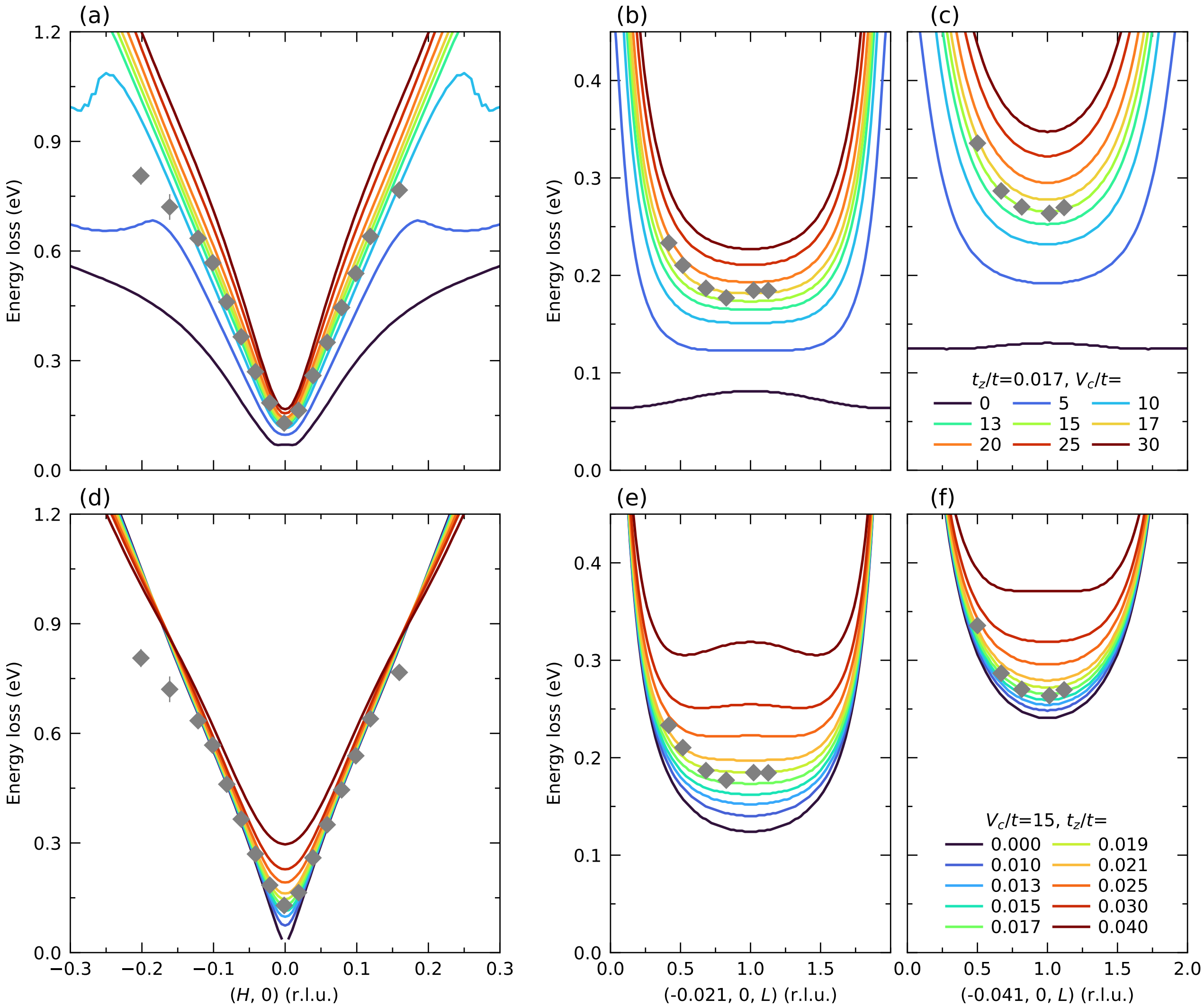

FIG. S4. Random phase approximation (RPA) calculations of $La_{2-x}Sr_xCuO_4$ ($\delta = 0.35$) plasmons as a function of tuning parameters. The diamonds are data points and the solid lines are calculation results. Based on the simulation results, the plasmon velocity for $V_c/t = 0$ [dark blue line in panel (a)] is approximately 1.8 eV Å.

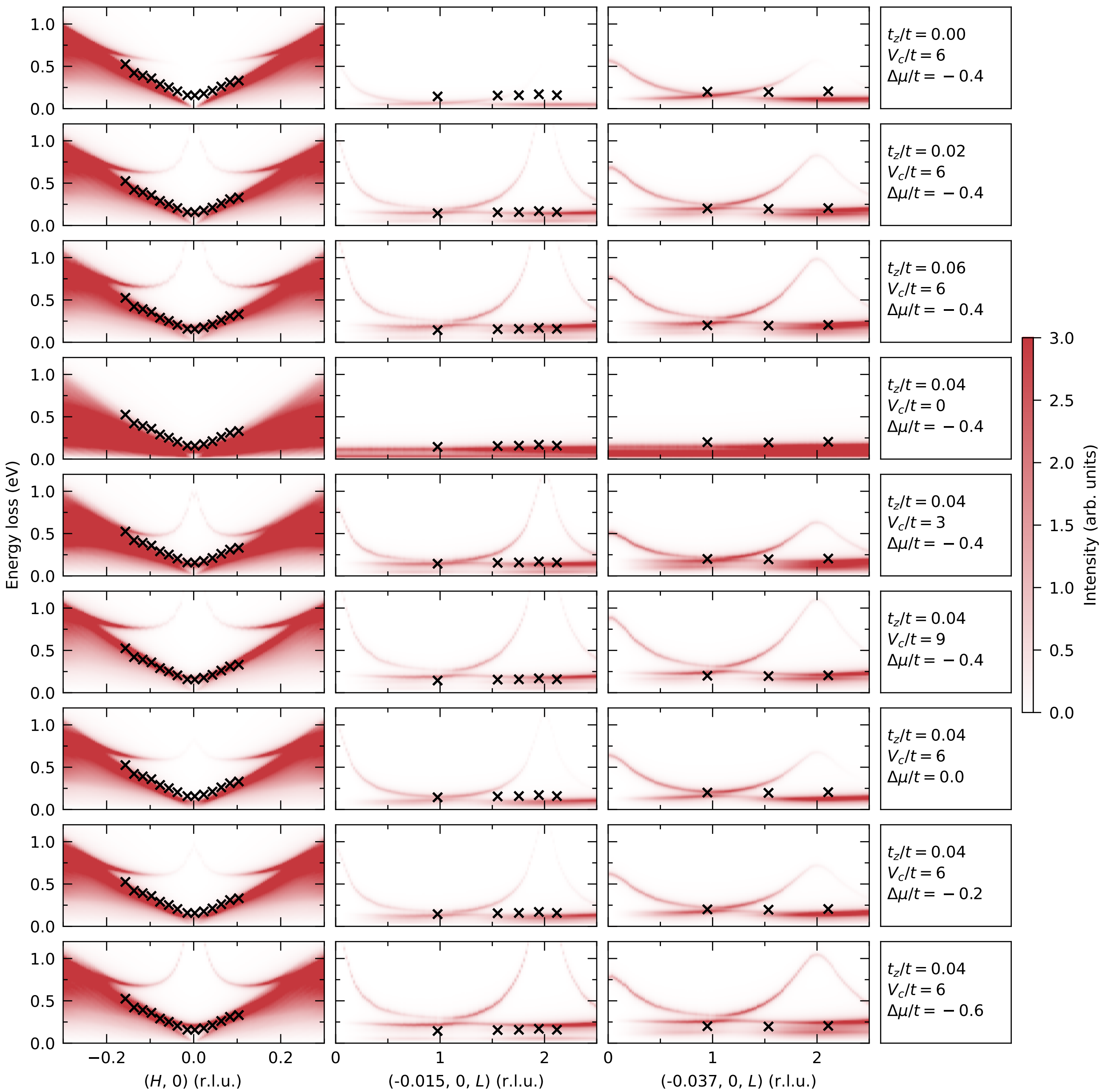

FIG. S5. RPA calculations of $Pr_4Ni_3O_8$ plasmons with tuning parameters. The crossings are data points.

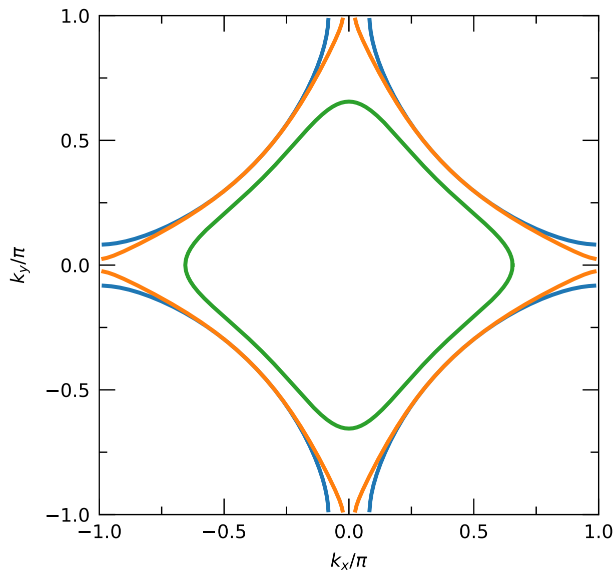

FIG. S6. Calculated $Pr_4Ni_3O_8$ Fermi surface using the parameters in the main text.

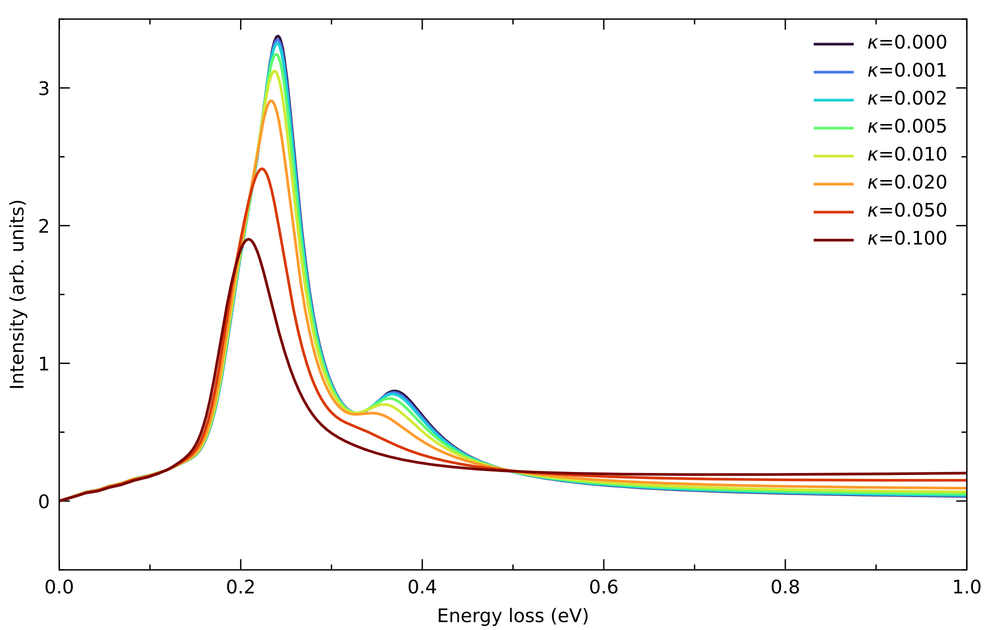

FIG. S7. RPA calculations of $Pr_4Ni_3O_8$ plasmons with various self-energy broadening.

[1] Junjie Zhang, AS Botana, JW Freeland, D Phelan, Hong Zheng, V Pardo, MR Norman, and JF Mitchell, "Large orbital polarization in a metallic square-planar nickelate," Nature Physics **13**, 864–869 (2017).

[2] Y. Shen, J. Sears, G. Fabbris, J. Li, J. Pelliciari, I. Jarrige, Xi He, I. Božović, M. Mitrano, Junjie Zhang, J. F. Mitchell, A. S. Botana, V. Bisogni, M. R. Norman, S. Johnston, and M. P. M. Dean, "Role of oxygen states in the low valence nickelate $La_4Ni_3O_8$," Physical Review X **12**, 011055 (2022).