

Supporting information for:

Understanding the different exciton-plasmon coupling
regimes in two-dimensional semiconductors coupled with
plasmonic lattices: a combined experimental and unified
equations of motion approach

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Number of Figures: 1 (Fig. S1)

Correlation between the equation of motion and the Hamiltonian matrix: As stated in the manuscript, the equation of motion for the coupled harmonic oscillators can be written as:

$$\ddot{x}_i + 2\gamma_i \dot{x}_i + \omega_i^2 x_i + 2 \sum_j \varepsilon_{ij} g_{ij} \dot{x}_j = \delta_{LSPR} E(t)$$

For steady state solution:

$$(\omega_i^2 + 2i\omega\gamma_i - \omega^2)x_i + 2i\omega \sum_j \varepsilon_{ij} g_{ij} x_j = \delta_{LSPR} E_0$$

We now apply the near-resonance condition $\omega \sim \omega_i$, and remove the driving force for now to solve only for the eigenstate problem, the equation is then simplified to:

$$(\omega_i + i\gamma_i - \omega)x_i + i \sum_j \varepsilon_{ij} g_{ij} x_j = 0$$

Rewriting the expression in the matrix form:

$$\begin{bmatrix} \omega_1 + i\gamma_1 - \omega & ig_{12} & ig_{13} & ig_{14} & ig_{15} \\ -ig_{12} & \omega_2 + i\gamma_2 - \omega & ig_{23} & ig_{24} & ig_{25} \\ -ig_{13} & -ig_{23} & \omega_3 + i\gamma_3 - \omega & ig_{34} & ig_{35} \\ -ig_{14} & -ig_{24} & -ig_{34} & \omega_4 + i\gamma_4 - \omega & ig_{45} \\ -ig_{15} & -ig_{25} & -ig_{35} & -ig_{45} & \omega_5 + i\gamma_5 - \omega \end{bmatrix} \begin{bmatrix} x_1 \\ x_2 \\ x_3 \\ x_4 \\ x_5 \end{bmatrix} = 0$$

By rearranging the matrix, we obtain:

$$\hat{H}\Psi = E\Psi$$

Where $E = \hbar\omega$ and \hat{H} stands for the Hamiltonian matrix of the system:

$$\hat{H} = \hbar \begin{bmatrix} \omega_1 + i\gamma_1 & ig_{12} & ig_{13} & ig_{14} & ig_{15} \\ -ig_{12} & \omega_2 + i\gamma_2 & ig_{23} & ig_{24} & ig_{25} \\ -ig_{13} & -ig_{23} & \omega_3 + i\gamma_3 & ig_{34} & ig_{35} \\ -ig_{14} & -ig_{24} & -ig_{34} & \omega_4 + i\gamma_4 & ig_{45} \\ -ig_{15} & -ig_{25} & -ig_{35} & -ig_{45} & \omega_5 + i\gamma_5 \end{bmatrix}$$

Hence by writing the equation of motion in the current form, the dimension and coefficient of the

coupling strength g_{ij} is consistent with the coupling strengths calculated from the Hamiltonian method.

In a two-level system, there is generally some freedom to choose the phase such that the coupling strength is real¹ without affecting the solution of the system's optical responses, e.g., absorption, scattering, emission, etc. In a mutually-coupled multi-level system such as the case in our system, of excitons coupled to a plasmonic lattice, however, the relative phases between the different coupling strengths can affect the dispersion spectra of the coupled system. In our study, we also examined the EOM fitting by changing the phases of the coupling strengths, and found that the best fit to the experiment was obtained when g_{ij} is real.

Figure S1. Validation of the near-resonance condition approximation (NRA)

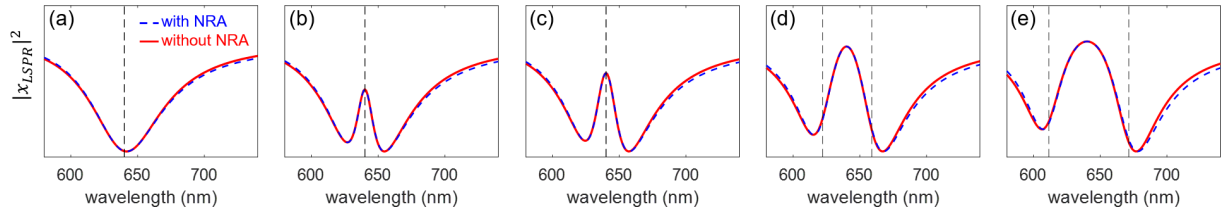


Figure S1 shows the calculated $|x_{LSPR}|^2$ spectra corresponding to Figure 4 (f)-(j) with and without the near-resonance condition approximation (NRA), where $\omega_0^2 - \omega^2$ is approximated as $2\omega(\omega_0 - \omega)$. Good correspondence between the two sets of simulated spectra indicates that this approximation is valid within the wavelength range of our study.

1. Walls, D. F.; Milburn, G. J., *Quantum optics*. Springer Science & Business Media: 2007.