Plasmon-exciton Interactions in Gold-WSe$_2$ Multilayer structures

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Abstract- Van der Waals materials such as thin films of transition-metal dichalcogenides (TMDCs) manifest strongly bound exciton states in the visible spectrum at ambient conditions that provide an ideal platform for exciton-photon couplings. Utilizing semiconducting TMDCs in the form of multilayer structure combined with metals can increase significantly the light-matter interaction. In this way, the interaction between excitons and surface-plasmon polaritons emerge as a platform for transferring the electromagnetic energy at confined modal volumes. Here, we theoretically investigate how moving electrons can be used as a probe of hybrid exciton-plasmon polaritons of gold-WSe$_2$ multilayers within the context of electron energy-loss spectroscopy (EELS) and cathodoluminescence (CL) spectroscopy. Interestingly, and in contrast to WSe$_2$ slab waveguides where quasi-propagating photonic modes interact with only exciton A, in gold-WSe$_2$ multilayer, exciton A and exciton B can both strongly interact with surface plasmon polaritons. Hence, we observe CL emission suppression at excitonic or plasmonic peaks, which reveals the energy transfer between excitons and plasmons in the form of nonradiating guided waves. Our work provides a systematic study for deeper understanding of the effect of the configuration and the thickness of layers on the photonic and plasmonic modes and hence on the strength of the coupling between excitons and surface-plasmon polaritons. Our findings pay the way for designing efficient photodetectors, sensitive sensors, and light emitting devices based on metal/semiconducting hybrid materials.
**Introduction**

Tailoring the interaction between light and matter is crucial for the performance of demanding optical and optoelectrical devices for applications ranging from sensing to computing [1-4]. Plasmonic [5-8] and high-index dielectric nanostructures [9-11] are frontier approaches for controlling light–matter interaction at the dimensions comparable to and smaller than the light wavelength. Semiconducting transition metal dichalcogenides (TMDCs) with their exciton-dominated optical response at room temperatures have recently emerged as a promising class of materials of realizing the strong coupling regime [12-14]. Among these excitonic materials, the TMDCs with Mo and W transition metals manifest themselves as high refractive-index materials, large oscillator strength, and large exciton binding energy [15-17]. Moreover, a sub-wavelength-thickness WSe$_2$ flake by itself reveals intriguingly a strong exciton-photon coupling [14, 18, 19]. However, the reduction of the optical mode volume as well as incorporating semiconducting TMDCs in more efficient photonic cavities, is one of the critical routes to further enhance and control the light-matter interaction strength [20]. Plasmon polaritons excitations in metals with their extraordinary ability to confine the electromagnetic energy below the diffraction limit are promising candidates to increase the light-matter coupling strengths in subwavelength dimensions. However, exploring the energy transfer dynamics and optical responses of hybrid Metal/TMDC structures demand subwavelength characterization techniques.

Electron-beam-based techniques such as electron energy-loss spectroscopy (EELS) [21-23] and cathodoluminescence (CL) spectroscopy [18, 24, 25] are normally applied to investigate the photonic responses of nanostructures. Recently, instrumental advancements in EELS performed in a scanning transmission electron microscope (STEM) allowed to spatially map phonon polaritons [26, 27] [28, 29], plasmon polaritons in [30-32], and localized plasmon [33-36]. A focused electron beam in a STEM can be used to probe the optical excitations in a broad spectral range and with a high spatial resolution. In addition to plasmon polaritons, recently exciton polaritons have been also experimentally investigated using electron beams [15, 18]. Plasmon-exciton polaritons (plexcitons) in metal/TMDC hybrid structures offer more versatility, enhanced light–matter interactions, faster carrier dynamics, and possibility to control the transport properties via plasmon-induced carrier doping [37-42].

To the best of our knowledge, the underlying mechanisms of the interaction of electron beams with multilayer structures remain largely unclear because the energy and the electromagnetic near-field associated with plasmon-exciton polaritons strongly depend on the geometry of the sample, the electron beam impact parameter, and the ordering of the layers in multilayered structure. Here, using a combination of theoretical
and numerical investigations, we demonstrate that both bulk and surface plasmons, as well excitons can be excited with electron beams. In addition, electron beams can probe plexciton modes that their resonant energies and dispersions strongly depend on the configuration of the multilayered structures. First, we present analytical calculation to explore exciton polaritons and plexcitons in planar semi-infinite multilayer structures. We further study the coupling between materials excitations in WSe$_2$ and Au to elucidate the effect of the structural geometry, such as their thicknesses and orderings on the coupling strength. Second, we study the formation of electromagnetic wave patterns in the field distributions induced by the electron beam in WSe$_2$ and Au hybrid structures. We also study the effect of lateral confinements caused by three-dimensional confinements. Our systematic investigations connect the excitation of these wake patterns with the different mechanisms caused by the moving electron traversing the structure such as excitation of exciton polaritons, surface plasmon polariton, plexcitons, and Cherenkov radiation.

Theoretical Analysis

When a swift electron with the charge of $q$ and the velocity of $v = \hat{z}v_e$ penetrates a multilayer structure, forward and backward radiation are generated at the interfaces, which is called transition radiation (TR). The origin of TR is the interaction of moving electron with its image charge as first predicted and further explained by Ginzburg and Frank [43]. TR, interestingly can be understood as generated by a uniformly moving electron beam and non-recoil approximation can be used to significantly simplify the theoretical treatments [44–46]. To calculate the intensity of the momentum-resolved electron energy-loss spectroscopy (MREELS) and CL of multilayer structures theoretically, we use a vector-potential approach [47]. To reach the optical response of multilayer structure theoretically, we consider an electron travelling parallel to $z$-axis. The first upper intersurface lies at the plane $z = 0$. The current density of a moving electron in the direction backward to the $z$-axis is expressed by $J_z(\vec{r}, t) = -e\nu_e \delta(x) \delta(y) \delta(z + \nu_z t)$. The vector potential $A_\alpha$ where $\alpha \in (x, y, z)$ is obtained by solving the inhomogeneous Helmholtz equation as (see Supplementary information for details)

$$A_\alpha = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dk_x dk_y \, e^{-ik_x x - ik_y y}$$

$$\left\{ \tilde{A}^{\alpha}_{n1} e^{-ik_z^{(n)}(z-z_n)} + \tilde{A}^{\alpha}_{n2} \, e^{ik_z^{(n)}(z+z_n)} + \delta_{\alpha z} \, e^{i\omega \nu_z - \frac{e\mu_0}{2\pi} - \frac{1}{\epsilon_r n k_0^2 - k_x^2 - k_y^2 - (\omega/v_e)^2}} \right\},$$

(1)

where $z_n$ defines the location of the upper and lower interfaces for the $n^{th}$ layer, $(k_z^{(n)})^2 = \epsilon_r n k_0^2 - k_x^2 - k_y^2$, and $\delta_{\alpha z}$ is the Kronecker-delta function, $k_0 = \omega/c$. $\tilde{A}^{\alpha}_{n1}$ and $\tilde{A}^{\alpha}_{n2}$ defines the unknown coefficients of the
plane waves propagating inside each layer in the upward and downward directions, respectively, that should be obtained using the boundary conditions. The index \( n \) spans the domain \([1, \ldots, N]\), where \( N \) is the number of the layers. Using the solution for the vector potentials, the electric field components are calculated as

\[
\vec{E}(\vec{r}, \omega) = -i\omega \vec{A}(\vec{r}, \omega) + \frac{1}{i\omega \varepsilon_0 \mu_0} \nabla \cdot \vec{A}(\vec{r}, \omega),
\]

and for the magnetic field we obtain

\[
\vec{H}(\vec{r}, \omega) = \nabla \times \vec{A}(\vec{r}, \omega)
\]

Using the Poynting’s theorem, the radiated power to the far field zone along the \( z \)-axis is recast as

\[
p_z = \frac{1}{2} Re \int dx \int dy \left(E_x H_y^* - E_y H_x^*\right) = \int dk_x \int dk_y \vec{p}_z(\omega; k_x, k_y).
\]

The EELS integral can be calculated as:

\[
\Gamma^{\text{EELS}} = \frac{-q}{2\pi \hbar \omega} Re \int \int_{-\infty}^{\infty} dk_x dk_y \int_{-\infty}^{\infty} dz \vec{E}_z(k_x, k_y, z, \omega) e^{-i\omega z/v_e}.
\]

and the MREEL spectrum is given by:

\[
\Gamma^{\text{EELS}}(\omega; k_x, k_y) = \frac{e}{2\pi \hbar \omega} \int_{-\infty}^{\infty} dz \vec{E}_z(k_x, k_y; z, \omega) e^{-i\omega z/v_e}.
\]

MREELS can be experimentally measured and can unambiguously resolve the dispersion diagram of polaritons. Hence, here we present the results of MREELS calculations to better understand exciton polaritons and plexcitons in metal/TMDC hybrid structures. After calculating MREELS for the bulk excitations as well as for radiated waves above and below the structure, and considering the guided waves within the films, we calculate different contributions to the EELS intensity (for more calculation details refer to supplementary section A).

**Results**

To explore the underlying physics, we begin by a WSe\(_2\) slab waveguide schematically showing in Fig. 1. In the followings, we first demonstrate the CL and EELS spectra of a thin flake of WSe\(_2\) structure. The permittivity of WSe\(_2\) shown in Fig. 1a demonstrates excitonic resonances at the energies of \( E = 1.68 \) eV and \( E = 2.1 \) eV, referred to as \( X^A \) and \( X^B \). As it is obvious from the MREELS patterns shown in Fig. 1d-f, because of the evanescent photonic modes below the cutoff range with large attenuation constant and short propagation lengths, in the 20nm-thick waveguide, only excitonic absorption peaks are apparent.
increasing the thickness of the waveguide, propagating photonic modes are excited and excitons strongly interact with photonic modes as shown in the 80-nm WSe$_2$ slab waveguide. In order to realize strong exciton-photon couplings, high-quality nano/microcavities [48-50], plasmonic nanoantennas [51-53], or plasmon-polariton nanosystems [54-56] are normally introduced. A strong interaction between cavity photons and excitons leads to the creation of exciton-polaritons. Recently, it has been demonstrated that the cavity-like Fabry-Pérot resonances result in strong exciton-photon interactions in freestanding WSe$_2$ planar waveguides [18]. Here, a moving electron with the kinetic energy of 30 keV traverses an 80-nm thick WSe$_2$ flake of (see Fig.1), which satisfies the Cherenkov radiation (CR) threshold ($\nu_{el} > c[\max(\epsilon_r)]^{-1/2}$), generates CR (CR) which strongly interacts with excitons in WSe$_2$[56-58]. Because the CR angle is larger than the critical angle of the total internal reflection in WSe$_2$ planar waveguide ($\frac{\epsilon_r \omega^2}{c^2} - \frac{\omega^2}{\nu^2} \frac{1}{c^2} > \frac{\omega^2}{c^2}$), the photons remain trapped inside the flakes and cannot contribute to the far-field radiation. Hence, the trapped CR causes standing-wave-like patterns of light and the anticrossing effect caused by strong exciton-photon interactions as shown in analytical and numerical results in Fig. 1b, f. Thus, we observe a deep in the CL spectra instead of the peak at the wavelength of 740 nm. To verify our analytical calculation, we have implemented numerical EELS and CL calculations in the commercial software COMSOL MULTIPHYSICS. As shown in Fig. 1b and c, our analytical and numerical results reveal a good agreement in the locations of peaks and deeps. Slight deviations from the analytical results happen due to the fact that in our numerical simulation domain, we have implemented a truncated slab waveguide with the width of $W = 2000$ nm. (see Supplementary A and Supplementary C sections for details of the analytical and the numerical methods, respectively).
Figure 1. (a) Real and imaginary parts of the relative permittivity of WSe$_2$ material. The dotted line marks A exciton and B excitons wavelengths. (b, c) Analytically (black line) and numerically (red dotted line) calculated cathodoluminescence and EELS intensities, respectively. The slab thickness is $d = 80$ nm. MREELS maps of WSe$_2$ slab waveguides with the thicknesses of (a) $d = 20$ nm, (b) $d = 60$ nm, (f) $d = 80$ nm. The exciton-photon coupling effect is limited due to the large attenuation and short propagation lengths for the photonic modes in the 20 nm-thick waveguide and therefore, only excitonic absorption peaks are observed. By increasing the thickness of the slab waveguide excitons start to interact with photons. The anticrossing behaviour is observed in the dispersion diagram of an 80-nm thick slab waveguide associated the emergence of bulk exciton-polaritons.

Here, we investigate in more details the CL and EELS spectra of a truncated WSe$_2$ slab (Fig. 2a). We observe a redshift in CL and EELS spectra by slightly changing the electron impact position from the edge of the slab waveguide (see Fig. 2(e, f)).
Figure 2. Schematic of the truncated slab waveguide. (a) Three-dimensional, (b) y-z plane views. The structure is a WSe$_2$ slab with the thickness of $d = 80$ nm and the width of $w = 2000$ nm surrounded by vacuum. An electron at the kinetic energy of $U = 30$ keV ($v_e = 0.223 \, c$) is moving along the negative $z$ direction at the distance $b$ from $x = 0$. (c) Cross-section of the total electric fields $|\vec{E}|$ calculated at $\lambda = 890$ nm for the on-edge electron trajectory ($b = w/2$) in the $x$-$z$ plane. (d) Same as in (c) but for the off-edge electron trajectory ($b = 0$). (e) Cathodoluminescence and (f) electron energy-loss spectrum versus the impact parameter $b$. Vertical dashed lines mark the A exciton and B exciton wavelengths for the bulk excitations. (g) and (h) Numerically calculated distance-wavelength CL and EELS probability spectra as a function of distance to the edge. Dotted green lines in EELS map trace the interference fringe maxima appearing due to reflection of the guided exciton polaritons from the slab edge.

By scanning the complete range from $b = 0$ to $W/2$ using the electron beam, we observe an interesting trend in the CL energy-distance map (marked by dashed black arrows), highlighting interference of transition radiation and diffraction radiation at far-field (Fig. 2g). The near-field interference effects in EELS energy-distance map are caused by the interference between propagating exciton polaritons and their reflection from the edges. In addition, due to the finite width of the slab in the direction perpendicular to the scanning direction, a second quantization effect causes the modulation of the interference fringes. The position of the emerging maxima is showed by dashed green lines. This reflection can be phenomenologically introduced into one dimensional Fourier transform of MREEL spectrum calculated by equation (6), by adding phase change of the propagation toward and back the edge as [59]:

$$\Gamma_{\text{EELS}}(\omega; y) = \int_0^\infty dk_\parallel \Gamma_{\text{EELS}}(\omega; k_\parallel) \left[ 1 + \cos \left( 2k_\parallel |y| + \varphi_{\text{ref}}(\omega) \right) \right]$$

where $2k_\parallel |x|$ is the propagation phase obtained by optical modes propagating toward and backward the edge. $\varphi_{\text{ref}}(\omega)$ is the phase imposed upon reflection from the edge that can be calculated for TM and TE modes by:

$$\varphi_{\text{TM}} = -2 \tan^{-1} \left( \frac{\cos^2 \theta_{ch} - \left( \frac{1}{\sqrt{\varepsilon_{\text{WSe}_2}}} \right)^2}{\sin \theta_{ch}} \right)$$

and for TE by:

$$\varphi_{\text{TE}} = -2 \tan^{-1} \left( \frac{\cos^2 \theta_{ch} - \left( \frac{1}{\sqrt{\varepsilon_{\text{WSe}_2}}} \right)^2}{\left( \frac{1}{\sqrt{\varepsilon_{\text{WSe}_2}}} \right)^2 \sin \theta_{ch}} \right)$$
where \( \theta_{ch} = \cos^{-1}\left(\frac{c}{\sqrt{\epsilon_{WSe_2} v_c}}\right) \) is Cherenkov angle.

Because only the TM mode can strongly interact with the excitons [18], we only consider equation (8) in calculation \( \varphi_{ref}(\omega) \) (for more details refer to supplementary section B). By plotting maximum of \( \Gamma^{EELS}(\omega; x) \) calculated according to Eq. (7), we find a good agreement between the results (dotted black line) and the fringe maxima obtained in numerical simulations, demonstrating the interference fringe maxima appearing due to reflection of the guided exciton polaritons from the slab edge.

Next, we demonstrate the simulated field profile associated with the electric field amplitude, for the same truncated slab waveguide and for two different electron impact positions, captured at the excitation wavelength of 890 nm. Obviously, we observe standing wave patterns caused by the Fabry-Perot resonances of the fundamental photonic modes inside the truncated waveguide. Fig. 2e and f show CL and EELS spectra at selected \( b \) parameters. Interestingly, we find two major differences with respect to the spectrum of an infinite WSe\(_2\) slab: (i) a red shift in the loss peak due to the guided mode excitation and its reflection from the edges (ii) shift in the CL peaks and deeps due to enhanced exciton-photon interactions in the truncated WSe\(_2\) slab waveguide.

In the next step, we explore the possibility of having exciton-plasmon interactions in multilayer Au/WSe\(_2\) geometries. The principal challenge of plasmonic is its non-switchable and passive response. Therefore, coupling between plasmons and exciton polaritons could be advantageous for active and nonlinear functionalities. As shown in schematic view of Fig. 3a and b, in WSe\(_2\) and Au multilayer structures, both exciton and plasmon polaritons can be excited by an electron beam traversing the structure. Because of high refractive index of WSe\(_2\), SPPs are mostly confined at the Au/WSe\(_2\) interface. Both plasmons and excitons can strongly interact with photons and create SPPs and exciton-polaritons, respectively. However, exciton polaritons can propagate in the bulk of the material in contrast with bulk plasmons. Since the permittivity of the WSe\(_2\) remains positive at the spectral range we consider here, only bulk exciton polaritons are excited, due to the strong interaction of the excitons with the propagating modes of the slab waveguide. Nevertheless, bulk exciton polaritons can also strongly interact with surface plasmons; this phenomenon is referred to as polariton-polariton interactions. We first consider a two-layer geometry composed of an 80-nm Au layer and a 150-nm WSe\(_2\) layer. Interestingly, the field profile of excited waves in this configuration depends on the propagation direction of the electron (Fig. 3b and c). By keeping the propagation direction of the electron along the negative \( z \)-axis and flipping the structure, we observe that the field profiles are not similar, thus
signifying that the time-ordering sequence of the exciton and plasmon polaritons excitations playing an important role. We will discuss this in more details later. Second, we consider two different three-layer configurations: (i) a 150-nm WSe$_2$ film sandwiched by two 80-nm gold films (Fig. 3d), and (ii) an 80-nm Au film sandwiched by two 150-nm WSe$_2$ films (Fig. 3e). In all cases, we observe a complicated wave front configuration. Particularly the wave front of both SPPs and exciton polaritons are disturbed and the distances between the maxima are not equidistance along the propagation detection. This signifies the coupling between excitons and plasmons and the emergence of plexcitons waves, which we discuss in more details in the followings.

In addition to field profiles, CL and EELS spectra of the two-layer structure strongly depend on the order of the layers (Fig. 4 a-f). In other words, the order on which the electron excites the plasmonic and excitonic responses, significantly alters the spectroscopy results. The fact the CL spectra demonstrate different peaks for both structures might be justified by the fact that we calculate the Ponting vector only towards the positive z-direction, thus calculating the reflected waves. However, EELs results demonstrate a different response for both structures as well. The comparison between analytical and numerical calculations show an overall good agreement, with the slight differences caused by the fact that for numerical calculations, we consider a truncated slab with the width of 2000 nm. In general, EELS and CL show different responses, and a shift of resonances, that we attribute to different mechanisms including (i) Stokes shift due to the contribution of loss channels to the absorption, and (ii) contribution of non-radiating polaritons, with their dispersion lines positioned outside of the light cone, to the EELS. Particularly the dispersion lines of these propagating polariton modes can be perfectly traced using MREELS calculations (Fig. 5a and b). Interestingly, we observe that SPP dispersion at the Au/vacuum surface is almost not affected by the presence of the WSe$_2$ layer, due to the 80-nm thickness of the Au layer, that avoids coupling between the SPPs at different interfaces. Nevertheless, the SPPs at the WSe$_2$/Au layers strongly interact with the exciton polaritons of the WSe$_2$ layer. Moreover, the resulting dispersion lines caused by the strong interaction depends on the way the electron excites the structure – a fact that we elaborate in more details in the followings.

Before discussing the underlying mechanisms for the strong-coupling effects in the two-layer structure, we discuss the results of the three-layer configurations. When two gold layers are positioned at a certain distance with respect to each other, void plasmons are excited in the vacuum between the layers, that acts as a more efficient cavity configurations compare to the total internal reflection at the boundaries of a WSe$_2$ layer. Void plasmons sustain a significantly lower attenuation constant compared to surface polaritons. Thus, placing a WSe$_2$ in between two Au layers, can results in a stronger exciton-photon interaction. A significant shift of
the peaks in both CL and EELS spectra compared to the exciton A, exciton B, and the plasmon peaks highlights the influence of strong coupling effects on the resulting eigen energies of the system (Fig. 4 g to i), as will be discussed later. The dispersion diagram of the new polaritons modes manifested in the MREELS maps (Fig. 5c) demonstrate that in this configuration, exciton B can also strongly interact with photons. In freestanding WSe₂ slab waveguide, because of faster relaxation rate and radiative nature of B-excitons in comparison to A-excitons [18], the interaction of photonic modes with A-excitons are much stronger and coupling of electrons with lower polariton branch are much pronounced, but by trapping the Cherenkov radiation in Au-WSe₂-Au, inside the sandwiched WSe₂ layer, the time of exciton-photon interaction increases, hence a strong coupling of B-excitons and photonic modes and anticrossing in upper polariton branch is observed. The enhancement of B excitons-photon coupling strength with increasing the WSe₂ thickness reveals that the higher order modes of multilayer structure responsible for the coupling of the electrons to upper polariton branch.

When the ordering of the layers is reversed, in the way that a gold layer at the thickness of 80 nm is sandwiched by two WSe₂ layers with the thickness of 150nm, the peaks of the CL spectra are only slightly shifted from the exciton A and B energies (Fig. 4 j and k), in contrast to the EELS spectrum (Fig. 4 l) The MREELS map mainly constitute lower polaritons branches excited at the energies below exciton A, and almost all of the EELS intensity is positioned outside the light cone, demonstrating a significant contribution of the interface SPPs and exciton polaritons to the EELS signal, compared to the radiation channels (Fig. 5d).
Figure 3. (a, b) Schematics of Au-WSe$_2$-Au and of WSe$_2$-Au-WSe$_2$ multilayer structure. Here, electrons can excite propagating plexcitons. The Thicknesses of Au and WSe$_2$ layers are $d_1 = 80$ nm and $d_2 = 150$ nm, respectively. (b-e) Cross section of the $z$-component of the induced electric field taken at $\lambda = 890$ nm for the on-edge electron trajectory in Au-WSe$_2$, WSe$_2$-Au, Au-WSe$_2$-Au, WSe$_2$-Au-WSe$_2$ multilayer structures, respectively. The plasmonic modes are transversely confined and longitudinally propagate along Au-WSe$_2$ interfaces.
Figure 4. (a, d, g, j) Structural schematic of gold-WSe$_2$, WSe$_2$-gold, gold-WSe$_2$-gold, and WSe$_2$-gold-WSe$_2$ multilayer structures, respectively, excited by an electron beam passing through the structures. The CL spectra is calculated at detector D$_1$ positioned at the distance of 700 nm above the structure. (b, e, h, k) Analytically calculated EEL$S$ and CL spectra. The Thicknesses of Au and WSe$_2$ layers are $d_1 = 80$ nm and $d_2 = 80$ nm, respectively. The exciton A, exciton B, and bulk-plasmon energies are indicated by dotted lines and labelled as P, $X^A$, and $X^B$, respectively. (c, f, i, l) The comparison of analytically and numerically calculated EEL$S$ spectra of (a, d, g, j) structures. Avoiding the repetition of the results, here, the thicknesses of Au and WSe$_2$ layers are chosen $d_1 = 150$ nm and $d_2 = 80$ nm, respectively.
In Au-WSe$_2$ multilayer structures, hybrid photonic modes not only strongly interact with A excitons, but also a strong interaction between photons and B excitons is observed. As we discussed previously, in the Au/WSe$_2$ two-layer structure, the ordering of the layers in the two-layer structure appearing along the electron trajectory can lead to the modification of the dispersion lines appearing in the MREELS map. Importantly, although the Cherenkov angle from the multilayer structure depends on the electron velocities, the created photons remain trapped by the total internal reflection at the dielectric interfaces and cannot contribute to radiation. Hence the Cherenkov radiation strongly couples to the plasmonic and excitonic excitations in the multilayer structure. Thus, the sequence the light interaction with the exciton and plasmon quasiparticles, is crucial and influences the electrodynamics response of the whole system. For gaining a deeper understanding, we use semi-classical model:

$$i \vec{P}_{ex} = (\omega_{ex}(k) - i \gamma) \vec{P}_{ex} + g \vec{P}_{pl} + I(t)$$

$$i \vec{P}_{pl} = (\omega_{pl}(k) - i \Gamma) \vec{P}_{pl} + g \vec{P}_{ex} + I(t - \tau)$$

(10)

Here, $\vec{P}_{ex}$ and $\vec{P}_{pl}$ are exciton and plasmon (-polariton) polarizations, respectively. $g$ is the coupling strength, $\omega_{ex}$ and $\omega_{pl}$ are the exciton and plasmon frequencies, respectively, and $\gamma$ and $\Gamma$ are the damping ratios associated with exciton and plasmon excitations, respectively. $I(t)$ is the time dependent excitation, that we assume to be specified by $I(t) = \delta(t)$. The magnitude of the delay is defined as $|\tau| = d_{Au}/v_{el}$, and is considered to be negative when the upper layer is the WSe$_2$ layer, and positive vice versa (for more information refer to supplementary Fig. 8 section B).

$$\vec{P}_{pl} = \frac{1}{\Delta}(g + (\omega - \omega_{ex}(k) + i \gamma)e^{-i\omega \tau})$$

$$\vec{P}_{ex} = \frac{1}{\Delta}(ge^{-i\omega \tau} + (\omega - \omega_{pl}(k) + i \Gamma))$$

(11)

where $\Delta = g^2 - (\omega - \omega_{pl}(k) + i \Gamma)(\omega - \omega_{ex}(k) + i \gamma)$
Figure 5. The energy-momentum EELS map of (a) Au-WSe$_2$, (b) WSe$_2$-Au, (c) Au-WSe$_2$-Au, (d) WSe$_2$-Au-WSe$_2$ multilayer structures. The electron at the kinetic energy of $U = 30$ keV interacts with photonic modes of the WSe$_2$-gold multilayer structures. The thicknesses of all layers are 80 nm. Only those photonic modes that contains an electric-field component oriented along the electron trajectory can interact with the probe electron. The coupling strength between excitons and SPPs increases, (the energy of gold plasmon completely transfer to exciton B of WSe$_2$ and there is a coupling between plasmon-exciton and exciton A of WSe$_2$. Confining the gold thin layer between two semiconductor layer or WSe$_2$ between two noble metal layers intensify the standing-wave-like patterns of light in the WSe$_2$ films.

Hence, in Au-WSe$_2$, WSe$_2$-Au, and Au-WSe$_2$-Au multilayer structure, the excited plasmon on air-Au interface and Au-WSe$_2$ interface can couple by the contribution of photonic modes. which leads plasmon-plasmon/plasmon-exciton photon mediated interactions. In the other word, in Au-WSe$_2$ and WSe$_2$-Au two-
layer structure, in thinner Au layer (less than 20 nm), the surface plasmon localized at the Au-WSe$_2$ interface decays exponentially across the Au film, and couples to the surface plasmon localized at air-Au interface. This occurs because the Au film is highly dissipative and carries more of the antisymmetric modes when the thickness decreases. By increasing the Au layer thickness to more than 40 nm, the SPP localized at Au-WSe$_2$ interface couples to radiation fields of air-Au interface (for more detail refer to Supplementary Figs. 5, 6). At 80 nm thickness, the Au film only supports one the localized modes between Au-WSe$_2$ interface. According to EELS map in Fig. 5, in Au-Wse$_2$-Au three-layer structure the radiation loss is small, because the photonic modes of air-Au interface lies on the nonradiative side of light line. However, in Au-Wse$_2$ and Wse$_2$-Au structures, the coupling of leaky modes of Au in Au-Wse$_2$ interface to excited surface plasmons and exciton-polaritons increase and leads to brighter results outside the light cone. By decreasing the Au layer thickness, the leaky modes and surface plasmon polariton and exciton-polaritons of WSe$_2$ increase, which leads to detection much more energy at far field.

Moreover, in the Au-WSe$_2$-Au configuration, it is possible to switch from forward Cherenkov radiation to the reverse radiation by decreasing the WSe$_2$ thickness below 55 nm [60]. The CL and EELS results of schematic view of WSe$_2$-Au-WSe$_2$ three-layer structure (schematic of Fig. 4j) reveals another fabulous phenomenon. Here, the energy of gold plasmon completely transfer to exciton B of WSe$_2$ and excitonic behaviour is the dominant response of the structure (see Fig. 4k and Fig. 5d). Recently, a giant anisotropy has reported for layered transition metal dichalcogenides such as WSe$_2$ which originates the fundamental differences between intralayer strong covalent bonding and weak interlayer van der Waals interaction [61]. According to our numerical simulations, this anisotropy doesn’t influence on the EELS and CL spectra of Au-WSe$_2$ multilayer structure so much and our calculations with assumption of isotropic dielectric function for WSe$_2$ is valid. This phenomenon was observed in previous researches [62].

**Discussion**

We have thoroughly precisely analyzed and simulate the excitation of optical exciton polaritons and surface exciton polariton in two-layer and three-layer Au-WSe$_2$ structure. We have observed when an electron travels through Au-WSe$_2$ multilayer structure, it can couple to excitons and plasmons of the structure and because the photons remain trapped at the dielectric-air interface, the CR stay inside waveguide cause Fabry-Pérot resonances which leads the anticrossing effect caused by strong light-matter interactions. we reveal the constructive interference of the transition radiation and the diffraction of the excited exciton-polaritons from the edges of the thin film flakes is a clear signature of the spontaneous coherence caused by the excitation of
exciton-polaritons. Here, we have shown in Au-WSe$_2$ multilayer, not only the confined photonic modes interact strongly with A excitons, but also strong interactions of photons and B excitons is observed. Also, by decreasing the thickness of Au layer, we observed plasmon-plasmon photon and plasmon-exciton coupling. We indeed demonstrate that the electromagnetic fields associated with excitation-polariton and plasmon-polaritons strongly depend on the geometry of the sample, the electron beam position, and the orientation of the stacked layers. Furthermore, by analytically and numerically studying the energy-momentum EELS and CL maps of Au-WSe$_2$ multilayer structure, we demonstrate that the order of the light interaction with matter are influential and can change the electrodynamics response of the structure. Our systematic findings can be generalized a guide for the correct interpretation light-matter interaction in excitonic and plasmonic multilayer structure.

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Supporting information

Plasmon-Exciton Interactions in Gold-WSe$_2$ Multilayer Structures

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Supplementary section A

The current density of a moving electron with the speed of $v_e$ along the z-axis is given by $J_z(\vec{r}, t) = -euv_e\delta(x)\delta(y)\delta(z - v_et)$. The vector potential ($\vec{A}$) is derived by solving the inhomogeneous Helmholtz equation $\vec{\nabla}^2 \vec{A}(\vec{r}, \omega) + \varepsilon_r(\omega)\mu_\epsilon k_0^2 \vec{A}(\vec{r}, \omega) = -\hat{z}\mu_0 \vec{J}_z(\vec{r}, \omega)$, $\vec{J}_z(\vec{r}, \omega)$ is the current density in frequency domain [2, 3].

The solution for the vector potential will be constructed as $\vec{A} = (A_x, 0, A_z)$, where the vector potential $A_\alpha$ can be obtained via solving the inhomogeneous Helmholtz equation for two-layer (Supplementary Figure 1) and three-layer configurations (Supplementary Figure 2) as:

Supplementary Figure 1: Schematic view of two-layer WSe$_2$-Au structure. The thicknesses of layers are $d_1$ and $d_2$. An electron at the speed of $u_e$ is moving along the negative z direction at the distance.
\[
\begin{align*}
A_\tau(x, y, z; \omega) &= \int_{-\infty}^{+\infty} dk_x \int_{-\infty}^{+\infty} dk_y \\
A_\sigma(x, y, z; \omega) &= \int_{-\infty}^{+\infty} dk_x \int_{-\infty}^{+\infty} dk_y \\
&
\begin{cases}
\tilde{A}^\alpha_1 \exp \left(-i k^{(1)}_z (z - z_1)\right) \exp \left(-i k_x x - i k_y y\right) + \delta_{\alpha z} \exp \left(i \frac{\omega}{v_e} z\right) \left(-\frac{e\mu_0}{2\pi} \right) \frac{1}{k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{v_e}\right)^2} & z > 0 \\
\tilde{A}^\alpha_{21} \exp \left(i k^{(2, \alpha)}_z (z - z_1)\right) \exp \left(-i k_x x - i k_y y\right) + \tilde{A}^\alpha_{22} \exp \left(-i k^{(2)}_z (z - z_2)\right) \exp \left(-i k_x x - i k_y y\right) + \delta_{\alpha z} \exp \left(i \frac{\omega}{v_e} z\right) \left(-\frac{e\mu_0}{2\pi} \right) \frac{1}{\varepsilon_r k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{v_e}\right)^2} & -d_1 < z \leq 0 \\
\tilde{A}^\alpha_{32} \exp \left(i k^{(3)}_z (z - z_2)\right) \exp \left(-i k_x x - i k_y y\right) + \tilde{A}^\alpha_{33} \exp \left(-i k^{(3)}_z (z - z_3)\right) \exp \left(-i k_x x - i k_y y\right) + \delta_{\alpha z} \exp \left(i \frac{\omega}{v_e} z\right) \left(-\frac{e\mu_0}{2\pi} \right) \frac{1}{\varepsilon_r k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{v_e}\right)^2} & -(d_1 + d_2) < z \leq -d_1 \\
\tilde{A}^\alpha_4 \exp \left(i k^{(1)}_z (z - z_3)\right) \exp \left(-i k_x x - i k_y y\right) + \delta_{\alpha z} \exp \left(i \frac{\omega}{v_e} z\right) \left(-\frac{e\mu_0}{2\pi} \right) \frac{1}{k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{v_e}\right)^2} & z \leq -(d_1 + d_2)
\end{cases}
\end{align*}
\] 

where \(k^{(1)}_z = k_0^2 - k_x^2 - k_y^2, (k^{(n)}_z)^2 = \varepsilon_r n k_0^2 - k_x^2 - k_y^2, \alpha \in (x, y, z), \) and \( n = 2, 3 \)

Upon the satisfying the boundary conditions of electric and magnetic fields, for unknown values are provided as:

\[
\begin{bmatrix} [M_{ij}]_{12 \times 12} \end{bmatrix}^{T} = \begin{bmatrix} [\tilde{I}]_{12 \times 1} \end{bmatrix}^{T}
\]

\[
\begin{bmatrix} [A] \end{bmatrix} = \begin{bmatrix} \tilde{A}^\alpha_1 \tilde{A}^\alpha_{21} \tilde{A}^\alpha_{22} \tilde{A}^\alpha_{32} \tilde{A}^\alpha_{33} \tilde{A}^\alpha_4 \tilde{A}^\alpha_{41} \tilde{A}^\alpha_{42} \tilde{A}^\alpha_{43} \tilde{A}^\alpha_{44} \end{bmatrix}
\]

\[
[I] = \begin{bmatrix} I_1 & I_2 & \ldots & I_{12} \end{bmatrix}
\]

Hence, the matrix elements are given by:

\[
\begin{align*}
m_{11}^x &= k_x k_z^{(1)}, m_{12}^x = k_x k_z^{(2)} / \varepsilon_{r_1}, m_{13}^x = m_{12}^x e^{-i k_z^{(2)} d_1}, m_{11}^z = -k_0^2 + k_x^2, m_{12}^z = \varepsilon_r k_0^2 - k_x^2 / \varepsilon_{r_1}, m_{13}^z = m_{12}^x e^{-i k_z^{(2)} d_1}, m_{21}^x = -k_x k_z^{(3)} / \varepsilon_{r_1}, m_{22}^x = m_{12}^x e^{-i k_z^{(2)} d_1}, m_{23}^x = -m_{22}^x e^{-i k_z^{(2)} d_1}, m_{24}^z = k_x k_z^{(3)} / \varepsilon_{r_1}, m_{25}^z = -m_{24}^x e^{-i k_z^{(2)} (d_2 - d_1)}, m_{22}^x = \varepsilon_r k_0^2 - k_x^2 / \varepsilon_{r_1} e^{-i k_z^{(2)} d_1}, m_{23}^z = m_{22}^x e^{-i k_z^{(2)} d_1}, m_{24}^z = \varepsilon_r k_0^2 - k_x^2 / \varepsilon_{r_2}, m_{25}^z = m_{24}^x e^{-i k_z^{(3)} (d_2 - d_1)}, m_{34}^z = -k_x k_z^{(3)} / \varepsilon_{r_2} e^{-i k_z^{(3)} (d_2 - d_1)}, m_{35}^z = m_{34}^x e^{-i k_z^{(3)} (d_2 - d_1)}, m_{41}^z = k_y k_z^{(1)} / \varepsilon_{r_2}, m_{42}^z = m_{41}^x / \varepsilon_{r_y,1}, m_{43}^x = m_{42}^x e^{-i k_z^{(2)} d_1}, m_{43}^z = m_{42}^x e^{-i k_z^{(2)} d_1}, m_{44}^x = -k_y k_z^{(2)} / \varepsilon_{r_1} e^{-i k_z^{(2)} d_1}, m_{52}^z = -k_y k_z^{(2)} / \varepsilon_{r_1} e^{-i k_z^{(2)} d_1}, m_{53}^z = m_{52}^x e^{-i k_z^{(2)} d_1}, m_{54}^x = k_y k_z^{(3)} / \varepsilon_{r_2} e^{-i k_z^{(3)} (d_2 - d_1)}, m_{55}^z = m_{54}^x e^{-i k_z^{(3)} (d_2 - d_1)}, m_{56}^z = k_y k_z^{(3)} / \varepsilon_{r_2}, m_{57}^x = m_{56}^x e^{-i k_z^{(3)} (d_2 - d_1)}, m_{58}^x = -k_y k_z^{(2)} / \varepsilon_{r_1} e^{-i k_z^{(2)} d_1}, m_{59}^x = m_{58}^x e^{-i k_z^{(3)} (d_2 - d_1)}, m_{61}^x = k_y / \varepsilon_{r_2}, m_{62}^x = m_{61}^x e^{-i k_z^{(3)} (d_2 - d_1)}, m_{63}^x = m_{62}^x e^{-i k_z^{(3)} (d_2 - d_1)}, m_{64}^x = -m_{63}^x e^{-i k_z^{(3)} (d_2 - d_1)}, m_{65}^x = -m_{64}^x e^{-i k_z^{(3)} (d_2 - d_1)}, m_{66}^x = -\varepsilon_{r_2} m_{65}^x, m_{71}^x = k_y, m_{72}^x = m_{71}^x e^{-i k_z^{(3)} d_1}, m_{81}^x = -k_x, m_{82}^x = m_{81}^x e^{-i k_z^{(3)} d_1}, m_{83}^x = m_{82}^x e^{-i k_z^{(3)} d_1}, m_{84}^x = m_{83}^x e^{-i k_z^{(3)} d_1}
\end{align*}
\]
\( m_{103} = m_{102} e^{-ik_x d_1}, \) \( m_{112} = m_{101} e^{-ik_x d_1}, \) \( m_{115} = -m_{101} e^{-ik_x (d_2-d_1)}, \) \( m_{112} = m_{102} e^{-ik_x d_1}, \) \( m_{114} = k_z, \)
\( m_{115} = -m_{114} e^{-ik_x (d_2-d_1)}, \) \( m_{124} = m_{101} e^{-ik_x (d_2-d_1)}, \) \( m_{124} = -m_{114} e^{-ik_x (d_2-d_1)}, \) and all the remaining undefined components are equal to 0.

The elements of \( I \) matrix are given by:

\[
I_1 = \frac{-e \mu_0 k_x}{2 \pi \epsilon_1 \left( \epsilon_{r_1} k_x^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e + \frac{e \mu_0 k_x}{2 \pi \epsilon_1 \left( k_x^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{i \frac{\omega}{\epsilon}_e d_1}
\]

\[
I_2 = \frac{-e \mu_0 k_x}{2 \pi \epsilon_2 \left( k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{-i \frac{\omega}{\epsilon}_e d_1} + \frac{e \mu_0 k_x}{2 \pi \epsilon_2 \left( k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{i \frac{\omega}{\epsilon}_e d_1}
\]

\[
I_3 = \frac{-e \mu_0 k_x}{2 \pi \left( k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{i \frac{\omega}{\epsilon}_e d_1} + \frac{e \mu_0 k_x}{2 \pi \epsilon_2 \left( k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{-i \frac{\omega}{\epsilon}_e d_1}
\]

\[
I_4 = \frac{-e \mu_0 k_y}{2 \pi \epsilon_1 \left( k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{i \frac{\omega}{\epsilon}_e d_1} + \frac{e \mu_0 k_y}{2 \pi \epsilon_1 \left( k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{-i \frac{\omega}{\epsilon}_e d_1}
\]

\[
I_5 = \frac{-e \mu_0 k_y}{2 \pi \epsilon_2 \left( \epsilon_{r_1} k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{-i \frac{\omega}{\epsilon}_e d_1} + \frac{e \mu_0 k_y}{2 \pi \epsilon_2 \left( \epsilon_{r_1} k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{i \frac{\omega}{\epsilon}_e d_1}
\]

\[
I_6 = \frac{-e \mu_0 k_y}{2 \pi \left( k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{-i \frac{\omega}{\epsilon}_e d_1} + \frac{e \mu_0 k_y}{2 \pi \epsilon_2 \left( k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{i \frac{\omega}{\epsilon}_e d_1}
\]

\[
I_7 = \frac{-e \mu_0 k_y}{2 \pi \epsilon_1 \left( \epsilon_{r_1} k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{-i \frac{\omega}{\epsilon}_e d_1} - \frac{e \mu_0 k_y}{2 \pi \epsilon_1 \left( \epsilon_{r_1} k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{i \frac{\omega}{\epsilon}_e d_1}
\]

\[
I_8 = \frac{e \mu_0 k_y}{2 \pi \epsilon_2 \left( \epsilon_{r_1} k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{-i \frac{\omega}{\epsilon}_e d_1} - \frac{e \mu_0 k_y}{2 \pi \epsilon_2 \left( \epsilon_{r_1} k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{i \frac{\omega}{\epsilon}_e d_1}
\]

\[
I_9 = \frac{-e \mu_0 k_y}{2 \pi \epsilon_2 \left( \epsilon_{r_1} k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{-i \frac{\omega}{\epsilon}_e d_1} - \frac{e \mu_0 k_y}{2 \pi \epsilon_2 \left( \epsilon_{r_1} k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{i \frac{\omega}{\epsilon}_e d_1}
\]

\[
I_{10} = \frac{-e \mu_0 k_x}{2 \pi \epsilon_1 \left( \epsilon_{r_1} k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{-i \frac{\omega}{\epsilon}_e d_1} - \frac{e \mu_0 k_x}{2 \pi \epsilon_1 \left( \epsilon_{r_1} k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{i \frac{\omega}{\epsilon}_e d_1}
\]

\[
I_{11} = \frac{e \mu_0 k_x}{2 \pi \epsilon_2 \left( \epsilon_{r_1} k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{-i \frac{\omega}{\epsilon}_e d_1} - \frac{e \mu_0 k_x}{2 \pi \epsilon_2 \left( \epsilon_{r_1} k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{i \frac{\omega}{\epsilon}_e d_1}
\]

\[
I_{12} = \frac{e \mu_0 k_x}{2 \pi \epsilon_2 \left( \epsilon_{r_1} k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{-i \frac{\omega}{\epsilon}_e d_1} - \frac{e \mu_0 k_x}{2 \pi \epsilon_2 \left( \epsilon_{r_1} k_0^2 - k_{x}^2 - k_y^2 - \left( \frac{\omega}{\epsilon}_e \right)^2 \right)} \frac{\omega}{\epsilon}_e e^{i \frac{\omega}{\epsilon}_e d_1}
\]
From the vector potential results, the electric field components can be calculated via:

$$\vec{E}(\vec{r}, \omega) = -i\omega \vec{A}(\vec{r}, \omega) + \frac{1}{i\omega \varepsilon_0 \mu_0} \vec{\nabla} \left( \vec{\nabla} \cdot \vec{A}(\vec{r}, \omega) \right)$$  \hspace{1cm} (2)

And for the magnetic field via:

$$\vec{H}(\vec{r}, \omega) = \nabla \times \vec{A}(\vec{r}, \omega)$$  \hspace{1cm} (3)

Using the pointing vector theorem, the radiated power to far field zone along z-axis is calculated by:

$$\vec{p}_z = \frac{1}{\mu_0} Re \int dx \int dy (E_x H_y^* - E_y H_x^*) = \int dk_x \int dk_y \vec{p}_z(\omega; k_x, k_y)$$  \hspace{1cm} (4)

The MREEL spectrum in diffraction can be defined as:

$$\Gamma_{\text{EELS}}^{\text{MREEL}}(\omega; k_x, k_y) = \frac{e^2}{2\pi \hbar \omega^2 \varepsilon_0} \frac{1}{\varepsilon_r} \frac{1}{\varepsilon_r} \frac{k_r^2 + k_z^2}{\varepsilon_r k_r^2 - k_z^2 - (\omega/\varepsilon_e)^2}$$  \hspace{1cm} (5)

To calculate the MREEL spectrum it is sufficient to calculate the z-component of electric field using eq. (5).

$$\Gamma_{\text{EELS}}^{\text{MREEL}}(\omega; k_x, k_y) = \Gamma_{\text{Bulk}}^{\text{EELS}}(\omega; k_x, k_y) + \Gamma_{\text{rad}}^{\text{EELS}}(\omega; k_x, k_y) + \Gamma_{\text{Guided}}^{\text{EELS}}(\omega; k_x, k_y)$$  \hspace{1cm} (6)

The bulk spectrum is calculated via:

$$\Gamma_{\text{Bulk}}^{\text{EELS}}(\omega; k_x, k_y) = \frac{e^2}{4\pi^2 \hbar \omega^2 \varepsilon_0} \frac{1}{\varepsilon_r} \frac{1}{\varepsilon_r} \frac{k_r^2 + k_z^2}{\varepsilon_r k_r^2 - k_z^2 - (\omega/\varepsilon_e)^2}$$  \hspace{1cm} (7)

Additionally, using the electric field in regions above and below the thin film, and considering the scattered electric field within the film, we calculate their contribution on EELS via:

$$\Gamma_{\text{rad, up}}^{\text{EELS}}(\omega; k_x, k_y) = \frac{e}{2\pi \hbar \omega^2 \varepsilon_0} \left\{ \frac{e^{-i(k_z^1/\varepsilon_e)}d_1}{\varepsilon_r k_r^2 - k_z^2 - (\omega/\varepsilon_e)^2} \left( k_r^1 k_x \bar{A}_4^1 + (k_0^2 - (k_z^1)^2) \bar{A}_4^1 \right) e^{-i(k_z^1)d_1} \right\} e^{-i(\omega/\varepsilon_e + k_z^1)d_1}$$  \hspace{1cm} (9)

$$\Gamma_{\text{rad, down}}^{\text{EELS}}(\omega; k_x, k_y) = \frac{e}{2\pi \hbar \omega^2 \varepsilon_0} \left\{ \frac{e^{-i(k_z^1/\varepsilon_e)}d_1}{\varepsilon_r k_r^2 - k_z^2 - (\omega/\varepsilon_e)^2} \left( -k_r^1 k_x \bar{A}_4^1 + (k_0^2 - (k_z^1)^2) \bar{A}_4^1 \right) e^{-i(k_z^1)d_1} \right\} e^{-i(\omega/\varepsilon_e + k_z^1)d_1}$$  \hspace{1cm} (10)

$$\Gamma_{\text{Guided}}^{\text{EELS}}(\omega; k_x, k_y) = \Gamma_{\text{rad, up}}^{\text{EELS}}(\omega; k_x, k_y) + \Gamma_{\text{Guided}}^{\text{EELS}}(\omega; k_x, k_y)$$  \hspace{1cm} (11)

$$\Gamma_{\text{Guided}}^{\text{EELS}}(\omega; k_x, k_y) = \frac{e^2}{2\pi \hbar \omega^2 \varepsilon_0} \left\{ \frac{\varepsilon_r k_r^2 - (k_z^2)^2}{\varepsilon_r^2} e^{-i(k_z^2)d_1} \times \left[ \bar{A}_2^1 \sin \left( k_z^2 \right) \right] + \bar{A}_2^2 \sin \left( k_z^2 + \omega/\varepsilon_e d_1 \right) \right\}$$  \hspace{1cm} (12)
\[
\frac{\omega}{v_e} d_2) + \bar{A}_{33}^x \text{sinc} \left( (k_z^{(3)} + \frac{\omega}{v_e}) d_2 \right) + \left( \frac{k_z k_z^{(3)}}{\varepsilon_{r_z}} \right) e^{-ik_z^{(3)} d_2} \times \left[ -\bar{A}_{31}^x \text{sinc} \left( \left( k_z - k_x \right) \right) \right]
\]

where \( k_z^{(1)} = k_0^2 - k_x^2 - k_y^2 \), \( k_z^{(2)} = \varepsilon_{r_1} k_0^2 - k_x^2 - k_y^2 \), \( k_z^{(3)} = \varepsilon_{r_2} k_0^2 - k_x^2 - k_y^2 \), and \( \alpha \in (x, y, z) \), and \( n = 2: 4 \).

Upon the satisfying the boundary conditions of electric and magnetic fields, for unknown values are provided as:

\[
\text{CL} = \frac{1}{8\pi^2 \mu_0 k_0} (k_x^2 k_z^{(1)} + |\bar{A}_{31}^x|^2 (k_x^2 + k_y^2) k_z^{(1)} - (\bar{A}_{11}^x \bar{A}_{11}^y + \bar{A}_{12}^x \bar{A}_{12}^y) k_z^{(1)} k_x)
\]

Supplementary Figure 2: Schematic view of three-layer WSe\(_2\)-Au structure. The thicknesses of layers are \( d_1 \) and \( d_2 \). An electron at the speed of \( v_e \) is moving along the negative \( z \) direction.

\[
A_{\alpha}(x, y, z; \omega) = \int_{-\infty}^{+\infty} dk_x \int_{-\infty}^{+\infty} dk_y
\]

\[
\begin{aligned}
&\bar{A}_{1\alpha}^x \exp \left( -ik_z^{(1)}(z - z_1) \right) \exp \left( -ik_x x - iky y \right) + \delta_{\alpha z} \exp \left( i \frac{\omega}{v_e} z \right) \frac{1}{\varepsilon_{r_1} k_0^2 - k_x^2 - k_y^2 - (\frac{\omega}{v_e})^2} & z > 0 \\
&\bar{A}_{21}^x \exp \left( ik_z^{(2\alpha)}(z - z_1) \right) \exp \left( -ik_x x - iky y \right) + \bar{A}_{22}^x \exp \left( -ik_z^{(2\alpha)}(z - z_2) \right) \exp \left( -ik_x x - iky y \right) + \delta_{\alpha z} \exp \left( i \frac{\omega}{v_e} z \right) \frac{1}{\varepsilon_{r_2} k_0^2 - k_x^2 - k_y^2 - (\frac{\omega}{v_e})^2} & -d_1 < z \leq 0 \\
&\bar{A}_{32}^x \exp \left( ik_z^{(3)}(z - z_2) \right) \exp \left( -ik_x x - iky y \right) + \bar{A}_{33}^x \exp \left( -ik_z^{(3)}(z - z_3) \right) \exp \left( -ik_x x - iky y \right) + \delta_{\alpha z} \exp \left( i \frac{\omega}{v_e} z \right) \frac{1}{\varepsilon_{r_3} k_0^2 - k_x^2 - k_y^2 - (\frac{\omega}{v_e})^2} & -(2d_1 + d_2) < z \leq -(d_1 + d_2) \\
&\bar{A}_{43}^x \exp \left( ik_z^{(4)}(z - z_3) \right) \exp \left( -ik_x x - iky y \right) + \bar{A}_{44}^x \exp \left( -ik_z^{(4)}(z - z_4) \right) \exp \left( -ik_x x - iky y \right) + \delta_{\alpha z} \exp \left( i \frac{\omega}{v_e} z \right) \frac{1}{\varepsilon_{r_4} k_0^2 - k_x^2 - k_y^2 - (\frac{\omega}{v_e})^2} & -(2d_1 + d_2) < z \leq -(d_1 + d_2) \\
&\bar{A}_{5}^x \exp \left( ik_z^{(1)}(z - z_4) \right) \exp \left( -ik_x x - iky y \right) + \delta_{\alpha z} \exp \left( i \frac{\omega}{v_e} z \right) \frac{1}{\varepsilon_{r_5} k_0^2 - k_x^2 - k_y^2 - (\frac{\omega}{v_e})^2} & z < -(2d_1 + d_2)
\end{aligned}
\]
And matrix elements are given by $m_{11} = k_x k_z (1)$, $m_{12} = k_x k_z (2)/\epsilon_r$, $m_{13} = -m_{22} e^{-i k_x (2) d_1}$, $m_{14} = -k_0 + k_x$, $m_{15} = \epsilon_r k_0 - \epsilon_r k_x /\epsilon_r$, $m_{22} = -k_x k_z (2)/\epsilon_r$, $m_{23} = -m_{22} e^{-i k_x (2) d_1}$, $m_{24} = k_x k_z (3)/\epsilon_r$, $m_{25} = -m_{24} e^{-i k_x (3) (d_2 - d_1)}$, $m_{26} = -\epsilon_r k_0 + k_x^2 /\epsilon_r$, $m_{27} = -m_{26} e^{-i k_x (2) d_1}$, $m_{28} = -k_y k_z (3)/\epsilon_r$, $m_{29} = -k_y k_z (4)/\epsilon_r$, $m_{32} = -k_0 e^{-i k_x (3) (d_2 - d_1)}$, $m_{33} = -k_0 e^{-i k_x (3) (d_2 - d_1)}$, $m_{34} = -k_y k_z (1)/\epsilon_r$, $m_{35} = -k_y k_z (2)/\epsilon_r$, $m_{36} = -k_y k_z (3)/\epsilon_r$, $m_{37} = -m_{36} e^{-i k_x (4) (d_1)}$, $m_{43} = -m_{46} e^{-i k_x (4) (d_1)}$, $m_{44} = k_y k_z (1)$, $m_{45} = k_y k_z (2)$, $m_{46} = e^{-i k_x (3) (d_2 - d_1)}$, $m_{47} = e^{-i k_x (3) (d_2 - d_1)}$, $m_{48} = \epsilon_r k_0 - k_x^2 /\epsilon_r$, $m_{53} = -m_{52} e^{-i k_x (2) (d_2 - d_1)}$, $m_{54} = m_{53} k_x /\epsilon_r$, $m_{55} = -m_{54} e^{-i k_x (2) (d_2 - d_1)}$, $m_{56} = m_{55} k_x /\epsilon_r$, $m_{57} = -m_{56} e^{-i k_x (3) (d_2 - d_1)}$, $m_{58} = -m_{57} e^{-i k_x (3) (d_2 - d_1)}$, $m_{62} = m_{63} k_x /\epsilon_r$, $m_{63} = m_{62} e^{-i k_x (3) (d_2 - d_1)}$, $m_{64} = m_{65} k_x /\epsilon_r$, $m_{65} = m_{64} e^{-i k_x (3) (d_2 - d_1)}$, $m_{66} = m_{65} k_x /\epsilon_r$, $m_{67} = -m_{66} e^{-i k_x (3) (d_2 - d_1)}$, $m_{68} = -m_{67} e^{-i k_x (3) (d_2 - d_1)}$, $m_{73} = k_x k_z (4)$, $m_{74} = m_{73} e^{-i k_x (4) (d_1)}$, $m_{75} = m_{74} e^{-i k_x (4) (d_1)}$, $m_{76} = k_y k_z (4)$, $m_{77} = m_{76} e^{-i k_x (4) (d_1)}$, $m_{78} = k_y k_z (4)$, $m_{83} = k_x k_z (4)$, $m_{84} = m_{83} e^{-i k_x (4) (d_1)}$, $m_{85} = m_{84} e^{-i k_x (4) (d_1)}$, $m_{86} = k_y k_z (4)$, $m_{87} = m_{86} e^{-i k_x (4) (d_1)}$, $m_{88} = m_{87} e^{-i k_x (4) (d_1)}$.

And for $I$ matrix we have:

$$l_1 = \frac{-\epsilon_0 k_x}{2\pi\epsilon_r (k_0^2 - k_x^2 - k_y^2)} \left( \frac{\omega}{\epsilon_r} \right)^2 \frac{2\pi\epsilon_r (k_0^2 - k_x^2 - k_y^2)}{\left( \frac{\omega}{\epsilon_r} \right)^2} \frac{2\pi\epsilon_r (k_0^2 - k_x^2 - k_y^2)}{\left( \frac{\omega}{\epsilon_r} \right)^2}$$

$$l_2 = \frac{-\epsilon_0 k_x}{2\pi\epsilon_r (k_0^2 - k_x^2 - k_y^2)} \left( \frac{\omega}{\epsilon_r} \right)^2 e^{-i \frac{\omega}{\epsilon_r} d_1} + \frac{2\pi\epsilon_r (k_0^2 - k_x^2 - k_y^2)}{\left( \frac{\omega}{\epsilon_r} \right)^2} \frac{2\pi\epsilon_r (k_0^2 - k_x^2 - k_y^2)}{\left( \frac{\omega}{\epsilon_r} \right)^2}$$

$$l_3 = \frac{-\epsilon_0 k_x}{2\pi\epsilon_r (k_0^2 - k_x^2 - k_y^2)} \left( \frac{\omega}{\epsilon_r} \right)^2 e^{-i \frac{\omega}{\epsilon_r} d_2} + \frac{2\pi\epsilon_r (k_0^2 - k_x^2 - k_y^2)}{\left( \frac{\omega}{\epsilon_r} \right)^2} \frac{2\pi\epsilon_r (k_0^2 - k_x^2 - k_y^2)}{\left( \frac{\omega}{\epsilon_r} \right)^2}$$

$$l_4 = \frac{-\epsilon_0 k_x}{2\pi\epsilon_r (k_0^2 - k_x^2 - k_y^2)} \left( \frac{\omega}{\epsilon_r} \right)^2 e^{-i \frac{\omega}{\epsilon_r} d_1} + \frac{2\pi\epsilon_r (k_0^2 - k_x^2 - k_y^2)}{\left( \frac{\omega}{\epsilon_r} \right)^2} \frac{2\pi\epsilon_r (k_0^2 - k_x^2 - k_y^2)}{\left( \frac{\omega}{\epsilon_r} \right)^2}$$
\[
I_5 = \frac{-e\mu_0k_y}{2\pi\varepsilon_r \left(\varepsilon_{r1} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) + \frac{e\mu_0k_y}{2\pi\varepsilon_r \left(\varepsilon_{r1} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right)
\]

\[
I_6 = \frac{-e\mu_0k_y}{2\pi\varepsilon_r \left(\varepsilon_{r2} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_1} + \frac{e\mu_0k_y}{2\pi\varepsilon_r \left(\varepsilon_{r2} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_2}
\]

\[
I_7 = \frac{-e\mu_0k_y}{2\pi\varepsilon_r \left(\varepsilon_{r2} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_1} + \frac{e\mu_0k_y}{2\pi\varepsilon_r \left(\varepsilon_{r2} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_2}
\]

\[
I_8 = \frac{-e\mu_0k_x}{2\pi\varepsilon_r \left(\varepsilon_{r1} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_1} + \frac{e\mu_0k_x}{2\pi\varepsilon_r \left(\varepsilon_{r1} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_1}
\]

\[
I_9 = \frac{e\mu_0k_y}{2\pi \left(\varepsilon_{r1} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_1} - \frac{e\mu_0k_y}{2\pi \left(\varepsilon_{r1} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_1}
\]

\[
I_{10} = \frac{e\mu_0k_y}{2\pi \left(\varepsilon_{r2} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_1} - \frac{e\mu_0k_y}{2\pi \left(\varepsilon_{r2} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_1}
\]

\[
I_{11} = \frac{e\mu_0k_y}{2\pi \left(\varepsilon_{r1} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_1} - \frac{e\mu_0k_y}{2\pi \left(\varepsilon_{r2} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_1}
\]

\[
I_{12} = \frac{e\mu_0k_y}{2\pi \left(\varepsilon_{r2} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_1} - \frac{e\mu_0k_y}{2\pi \left(\varepsilon_{r2} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_1}
\]

\[
I_{13} = \frac{-e\mu_0k_x}{2\pi \left(\varepsilon_{r1} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) + \frac{e\mu_0k_x}{2\pi \left(\varepsilon_{r1} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right)
\]

\[
I_{14} = \frac{-e\mu_0k_x}{2\pi \left(\varepsilon_{r2} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_1} + \frac{e\mu_0k_x}{2\pi \left(\varepsilon_{r2} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_1}
\]

\[
I_{15} = \frac{-e\mu_0k_x}{2\pi \left(\varepsilon_{r1} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_2} + \frac{e\mu_0k_x}{2\pi \left(\varepsilon_{r2} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_2}
\]

\[
I_{16} = \frac{-e\mu_0k_x}{2\pi \left(\varepsilon_{r2} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_1} + \frac{e\mu_0k_x}{2\pi \left(\varepsilon_{r1} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2\right)} \left(\frac{\omega}{\nu_e}\right) e^{-i\frac{\omega}{\nu_e}d_1}
\]

To calculate the MREEL spectrum it is sufficient to calculate the z-component of electric field using eq. (5-7).

\[
\Gamma_{\text{EELS}}^{\text{Bulk}}(\omega; k_x, k_y) = \frac{e^2}{4\pi^2 \hbar \omega^2 \varepsilon_0} \left(2d_1 \frac{\varepsilon_{r1} k_0^2 - \left(\frac{\omega}{\nu_e}\right)^2}{\varepsilon_{r1} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2} + d_2 \frac{\varepsilon_{r2} k_0^2 - \left(\frac{\omega}{\nu_e}\right)^2}{\varepsilon_{r2} k_0^2 - k_x^2 - k_y^2 - \left(\frac{\omega}{\nu_e}\right)^2}\right) \quad (14)
\]
\[ \Gamma_{\text{rad up}}(\omega; k_x, k_y) = \frac{e^{i\omega \frac{\varepsilon_0}{v_e}d}}{2\pi \varepsilon_0 \varepsilon_0} \left( e^{-i\left(\frac{\omega}{v_e}\right)d} \left( k_x^{(1)} k_x \hat{A}_1^x + \left( k_0^2 - (k_x^{(1)})^2 \right) \hat{A}_1^x \right) e^{-i(k_z^{(1)})d_1} \right) e^{-i\left(\frac{\omega}{v_e}+k_z^{(1)}\right)d_1} \] (15)

\[ \Gamma_{\text{rad down}}(\omega; k_x, k_y) = \frac{e^{i\omega \frac{\varepsilon_0}{v_e}d}}{2\pi \varepsilon_0 \varepsilon_0} \left( e^{-i\left(\frac{\omega}{v_e}\right)d} \left( -k_x^{(1)} k_x \hat{A}_5^x + \left( k_0^2 - (k_z^{(1)})^2 \right) \hat{A}_5^x \right) e^{-i\left(\frac{\omega}{v_e}+k_z^{(1)}\right)d_2} \right) \] (16)

\[ \Gamma_{\text{rad}}(\omega; k_x, k_y) = \Gamma_{\text{rad up}}(\omega; k_x, k_y) + \Gamma_{\text{rad down}}(\omega; k_x, k_y) \] (17)

\[ \Gamma_{\text{Guided}}(\omega; k_x, k_y) = \frac{e^{i\omega \frac{\varepsilon_0}{v_e}d}}{2\pi \varepsilon_0 \varepsilon_0} \left( \frac{\varepsilon_{r1d1} k_0^2 (k_z^{(2)})^2}{\varepsilon_{r1}} e^{-i\frac{k_z^{(2)}}{d_1}d_1} \right) \left[ \hat{A}_{21}^x \text{sinc} \left( k_z^{(2)} \frac{\omega}{v_e} d_1 \right) + \hat{A}_{22}^x \text{sinc} \left( k_z^{(2)} + \frac{\omega}{v_e} d_1 \right) \right] + \left( k_x k_z^{(2)} \right) \frac{\varepsilon_{r2d2} k_0^2 (k_z^{(3)})^2}{\varepsilon_{r2}} e^{-i\frac{k_z^{(3)}}{d_2}d_2} \left[ \hat{A}_{31}^x \text{sinc} \left( k_z^{(3)} - \frac{\omega}{v_e} d_2 \right) + \hat{A}_{33}^x \text{sinc} \left( k_z^{(3)} + \frac{\omega}{v_e} d_2 \right) \right] \] + \left( k_x k_z^{(4)} \right) \frac{\varepsilon_{r4d4} k_0^2 (k_z^{(4)})^2}{\varepsilon_{r4}} e^{-i\frac{k_z^{(4)}}{d_4}d_4} \left[ \hat{A}_{43}^x \text{sinc} \left( k_z^{(4)} - \frac{\omega}{v_e} d_4 \right) + \hat{A}_{44}^x \text{sinc} \left( k_z^{(4)} + \frac{\omega}{v_e} d_4 \right) \right] \] (18)
Supplementary Figure 3: Calculated MREELS for thick infinite (a) and (b) truncated WSe$_2$ slab waveguides with the width of $w = 2000 \text{nm}$ for the thickness of $d = 300 \text{nm}$. Comprising EELS map of (a) and (b), in the analytically calculated EELS map of infinite WSe$_2$ slab waveguide in (a), we have three propagation photonic modes which is the results of the interaction Cherenkov radiation (CR) with excitons of WSe$_2$ which leads to exciton-polariton propagations but, the numerically calculated EELS map of truncated WSe$_2$ slab waveguide in (b) reveals the propagation and the reflection of photonic modes in the longitudinal and the transversal directions which lead more photonic modes. The confinement of standing-wave-like patterns of the trapped in two dimensions enhance the exciton-photon interactions. Hence, the energy splitting and anticrossing in the energy-momentum relation of the emergent exciton-polaritons in (b) is more intensive than (a). Moreover, in infinite WSe$_2$ waveguide (a), because the CR angle is larger than the critical angle of the total internal reflection the trapped photons cannot contribute to radiation in far field (above the white dash lines represent photon dispersion in free space) but in truncated slab waveguide(b) the confined energy can escape from the edges due to the interaction of the electron with edges (diffraction radiation) and the scattering of exciton-polaritons from edges can be detected much brighter in far field.
Supplementary Figure 4: Calculated MREELS for depicted thicknesses of WSe$_2$ and Au layers in Au-WSe$_2$-Au multilayer structure. The Bulk EELS map has demonstrated an energy exchange between the bulk plasmon of gold and B exciton-polariton of WSe$_2$ in. The enhancement of plasmon-plasmon and plasmon-exciton coupling with decreasing the Au thickness is traceable from (a) to (c). as demonstrated in (a), the excited plasmon on air-Au interface and Au-WSe$_2$ interface can couple by the contribution of photonic modes, which leads to the plasmon-plasmon/plasmon-exciton photon mediated interactions, these phenomena are much pronounced in two-layer Au-WSe$_2$ and WSe$_2$-Au as shown in Supplementary Figure 5 and 6.
Supplementary Figure 5: Calculated MREELS for depicted thicknesses of WSe_2 and Au layers in Au-WSe_2 multilayer structure. The enhancement of plasmon-plasmon/plasmon-exciton photon mediated interactions strength between the excited plasmons of air-Au, Au-WSe_2 interfaces, and excitons of WSe_2 by decreasing the Au thickness can be observed in (a) and (b).

Supplementary Figure 6: Calculated MREELS for depicted thicknesses of WSe_2 and Au layers in WSe_2-Au multilayer structure. The enhancement of plasmon-plasmon strength coupling between the excited plasmons of air-Au and Au-WSe_2 interfaces by decreasing the Au thickness can be observed in (a) and (b).
Supplementary Figure 7: Calculated MREELS for depicted thicknesses of WSe$_2$ and Au layers in Au-WSe$_2$-Au multilayer structure. The enhancement of B excitons-photon coupling strength with increasing the WSe$_2$ thickness is traceable from (a) to (b). The interaction of photonic modes with A-excitons are much stronger and coupling of electrons with lower polariton branch, but by trapping the Cherenkov radiation in Au-WSe$_2$-Au, inside the sandwiched WSe$_2$ layer, the time of exciton-photon interaction increases, the anticrossing in upper polariton branch is observed. This is because of the strong coupling of B-excitons and photonic modes. The enhancement of B excitons-photon coupling strength with increasing the WSe$_2$ thickness reveals that the higher order modes of multilayer structure responsible for the coupling of the electrons to upper polariton branch. Also, in comparison with free-standing WSe$_2$ waveguide (See figure 1), here, in the thicknesses less than 60 nm we can observe lower branch exciton-photon interaction but in free-standing WSe$_2$ slab, exciton-photon coupling effect is limited due to the large attenuation and short propagation lengths for the photonic modes, only excitonic absorption peaks are observed.
Supplementary Figure 8: Calculated (a) A exciton-plasmon (b) B exciton-plasmon (-polariton) polarizations coupling based on delay (Au layer thickness) in various energies. For plasmon-A excitons interaction, the minimum delay to observe significant change in plasmon polarization in at negative delay region (WSe$_2$-Au ordering) is 0.5 fs which related to 82.17 nm thickness of Au layer, and in positive region (Au-WSe$_2$ region) is 0.55 fs which related to 90.4 nm thickness of Au layer at the energy of exciton A. As demonstrated in (a) and (b) the exciton-plasmon energy at energy of plasmon (near 2.4 eV) is much sensitive to the change of Au layer and it experiences much more changes by changing the Au layer thickness. In plasmon-B excitons interaction, the minimum delay to observe significant change in plasmon polarization in at negative delay region (WSe$_2$-Au ordering) is 0.2 fs which related to 32.8 nm thickness of Au layer, and in positive region (Au-WSe$_2$ region) is 1.15 fs which related to 189 nm thickness of Au layer.
Supplementary Figure 9: Calculated EELS for various thicknesses of Au and WSe$_2$ layers in (a) WSe$_2$-Au-WSe$_2$ and (b) Au-WSe$_2$-Au multilayer structure, respectively. Dash black lines trace the maxima of interacting resonances of the excitons and photonic modes appearing due interactions of the exciton polaritons of three-layer structures. The dotted line marks A exciton and B excitons and bulk plasmons wavelengths. By decreasing the thickness of sandwiched layer (Au in (a) and WSe$_2$ in (b)), the light-matter strongly interactions increase. As demonstrated here, the interactions of photonic modes, plasmons and excitons grow dramatically for the thicknesses of below 40nm which is in good agreement with the results of supplementary Figs. 4 and 5. The thicknesses of WSe$_2$ layers in WSe$_2$-Au-WSe$_2$ and Au layers in Au-WSe$_2$-Au structures are 80 nm.

We can calculate the phase shift experienced by the optical waves upon reflection from an interface based on the theory proposed by Artmann [4]. When a moving electron with the kinetic energy of 30 keV traverses a WSe$_2$ slab waveguide, the angle of the generated Cherenkov radiation (CR) is larger than the critical angle of the total internal reflection in WSe$_2$ planar waveguides. Therefore, the photons remain trapped inside the flakes and causes standing-wave-like patterns of light. By assuming propagating waves along the $y$ direction, the wavevector is given by:

$$k_y = \frac{2\pi}{\lambda} \sin \theta$$  \hspace{1cm} (1)

The wave is represented by $\exp(ik_y y)$. There is a phase shift at the edge, hence the reflected wave is given by $\exp(ik_y y + \varphi)$. The superposition of forward and backward waves is given by:

$$\psi = (\exp(ik_y y + \varphi) + \exp(i[k_y + \Delta k_y]y + \Delta \varphi))$$ \hspace{1cm} (2)

The maxima lie on:

$$\Delta k_y y + \Delta \varphi = 2\pi n$$ \hspace{1cm} (3)

Where $n$ is a whole number.

On the other hand, if the reflection process happens from a perfect metal surface, equation (3) is changed to:

$$\Delta k_y y_{metal} = 2\pi n$$ \hspace{1cm} (4)
By subtracting equations 3 and 4, we can obtain the expression of the phase shift:

$$\theta = \frac{\Delta \phi}{\Delta k_y}$$  

(5)

According to Maxwell theory the phase change of reflection for TM and TE modes are given by:

$$\varphi_{TM} = -2\tan^{-1}\left(\frac{\sin^2 \theta - \left(\frac{1}{\sqrt{\varepsilon_{\text{WS}e_2}}}\right)^2}{\cos \theta}\right)$$  

(6)

$$\varphi_{TE} = -2\tan^{-1}\left(\frac{\left(\frac{1}{\sqrt{\varepsilon_{\text{WS}e_2}}}\right)^2 - \sin^2 \theta}{\cos \theta}\right)$$  

(7)

Where $\theta$ is equal to $\frac{\pi}{2} - \theta_{\text{cherenkov}}$.

Supplementary Figure 10: $\varphi_{\text{ref}(\omega)}$, the phase imposed upon reflection from the edge calculated by assuming TM modes in a truncated WSe$_2$ slab waveguide.
Supplementary Figure 10: $\phi_{\text{ref}}(\omega)$, the phase imposed upon reflection from the edge, calculated by assuming TE modes in a truncated WSe$_2$ slab waveguide.
Supplementary section C

We have employed both a home-built analytical solution code and the COMSOL Multiphysics software to gain insight into the temporal distribution of the electron-induced radiation, exciton-photon, and plasmons interactions in our Au-WSe$_2$ multilayer structures. As discussed in Supplementary section A, to calculate the intensities of momentum-resolved electron energy-loss spectroscopy (MREELS) and CL of multilayer structure theoretically, we use a vector-potential approach. We model an electron beam by a current density distribution corresponding to a swift electron at a kinetic energy of $U = 30$ keV. To calculate EELS and CL data in the frequency domain using a finite element method, we have employed the radiofrequency (RF) toolbox of COMSOL in a 3D simulation domain, which is based on solving the Maxwell equations in real space and in the frequency domain. We have utilized an oscillating “edge current” as a fast electron beam along a straight line representing the electron beam. The current was expressed by $I = I_0 \exp(i\omega z/v_e)$. The moving electron (represented by a linear current) acts as a source, creating the electromagnetic field in the system. The CL signal was calculated by using a boundary probe at a plane normal to the electron beam. The dielectric function of WSe$_2$ and Au were defined by using the interpolation function of COMSOL. We apply the free tetrahedral mesh with refined elements close to the electron’s trajectory. The area of PML is meshed by 15 Swept layers. Because the Geometry of the multilayered structure, Perfectly Matched Layers (PML) with Cartesian symmetry help to attenuate the electric field at the boundaries of the simulation domain and prevent unphysical field reflections from the boundaries.

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