Enhanced coherent interaction between monolayer WS$_2$ and film-coupled nanocube open cavity with suppressed incoherent damping pathway

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Future quantum information devices will most likely rely on the realization of coherent light-matter interactions in strong coupling regime and the maintaining of the coherence with minimized incoherent damping pathways. Here, utilizing film-coupled planar nanoparticle supporting unique plasmon-induced magnetic resonance (PIMR), we theoretically study a strong coupling of a single nanocube to a monolayer of two-dimensional atomic crystal. We demonstrate that the nanocube-based planar configuration with magnetic flux passing through the dielectric layer exhibits much higher in-plane field confinement with respect to traditional plasmon electric modes, leading to an efficient coherent coupling to only a few in-plane excitonic dipoles with a record of normal mode splitting over 280 meV. Importantly, a much reduced incoherent coupling strength down to 3 meV is obtained. We reveal that the underlying mechanism lies in two main facts: (i) the small radiative damping rate of the excitonic system giving very limited contribution to the linewidth broadening of the A-exciton resonance, and (ii) the excitation of magnetic resonance provides a net electric dipole moment orientating mainly normal to the film surface, thus greatly suppressing the exchange of photons between the two subsystems via the continuum reservoir. The numerical simulations and theoretical analysis quantitatively evaluate the dependence of both the coherent and incoherent coupling strength on the thickness of the dielectric layer, revealing the fact that coherent/incoherent coupling strength can be simultaneously enhanced/suppressed with the decrease/increase of the film thickness, which can be appropriately designed to achieve the optimal coherent coupling with minimized incoherent coupling strength. Such hybrid nanostructure with simple geometry and ease of fabrication may not only offer as an attractive platform to explore light-matter interaction in the strong coupling regime but also show potential applications in realizing novel quantum and nanophotonic optical devices.

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I. INTRODUCTION

Strong coupling of electronic transitions to plasmonic nanocavities supporting surface plasmons (SPs) has inspired tremendous studies on light-matter interactions at the nanoscale for its fundamental importance both in cavity quantum electrodynamics and in quantum information applications [1,2]. Strong coupling occurs when the coupling strength between the two subsystems exceeds their decoherence or dissipation rates, resulting in the formation of hybrid states with mixed light-matter features [3–7]. The key step for reaching strong coupling relies on increasing the coupling strength while reducing the damping rates of the subsystems. The coupling strength is typically determined as $g = \sqrt{N} \mu_e \cdot E \propto \mu_e \sqrt{N/V}$ and proportional to the inner product of the dipole moment $\mu_e$ of the quantum emitter and the electric field $E$ confined within the mode volume $V$ [4,5]. Here, $N$ is the number of emitters contributing to the coherent coupling. In this sense, strong coupling between excitons in organic/inorganic semiconductors and plasmonic nanocavities offered by metallic nanoparticles has been widely studied due to the SPs with deeply subwavelength mode confinement and large dipole moments of the excitonic transitions [8–18].

Two-dimensional (2D) transition-metal dichalcogenides (TMDs) [19] are emerging as a new promising type of materials for the study of light-matter coupling due to their unique advantages over organic semiconductors, such as high transition dipole moments, atomic thickness, chemical inertness, and optical stability. Special interest has been drawn in the strong coupling between a single layer of TMDs and nanocavities formed by single plasmonic nanoparticles (NPs) [20–23], particularly by single NPs on metal film separated by a dielectric nanospacer, typically called NPs on mirror (NPoM) owing to their ability to confine the optical modes within an ultrasmall mode volume [24]. Recent studies have demonstrated strong coupling of TMDs with NPoM systems containing a different shape of single metallic NPs [25–28]. In a NPoM system with a single gold nanosphere coupling to seven-layer WS$_2$, coherent coupling between WS$_2$ excitons and the gap-mode plasmons with a Rabi splitting over 135 meV was achieved at room temperature [25].
Very recently, we have demonstrated an enhanced strong coupling with a Rabi splitting over 200 meV in a coupling system consisting of single layer TMDs and magnetic resonances resulting from plasmon-hybridization effects, known as plasmon-induced magnetic resonance (PIMR) [29–31] generated in a NPoM system with a single silver nanorod [32]. The PIMR is excited by the displacement current loop induced by the coupling among different plasmon modes, leading to strong magnetic and electric “hot spots” with much improved local field enhancement with respect to transitional electrically excited plasmons [31]. In our previous NPoM system with single silver nanorod, the large coherent coupling is accompanied by a considerable incoherent damping process originating from the mutual cross damping pathway via the continuum reservoir, which introduces an inevitable decoherence to the coherent interaction process and therefore hinders their potential applications in quantum information processing and quantum entanglement where the strong coherent light-matter coupling with the persistence of coherence is highly desired.

In this paper, we demonstrate a strong coherent coupling of a NPoM system with a monolayer of WS2, exhibiting a much improved coherent strong strength with a drastic suppression of incoherent damping process. By employing a NPoM structure with a silver nanocube over a silver substrate separated by a TiO2 dielectric layer, a record of Rabi splitting over 280 meV is obtained with a minimum coupling-induced incoherent coupling strength down to 3 meV. Detailed simulations and theoretical analysis reveal the physical mechanism behind the enhanced coherent coupling strength and much reduced incoherent damping process. We also quantitatively evaluate the dependence of both the coherent and incoherent coupling strength on the thickness of the dielectric layer, demonstrating the possibility of controlling both the coherent and incoherent coupling processes simultaneously for the optimal coherent coupling with minimized incoherent coupling strength.

II. RESULTS AND DISCUSSION

A. Optical response of the hybrid system

The proposed nanostructure, as demonstrated in Fig. 1(a), consists of a silver nanocube deposited on a silver substrate, which is coated with a thin dielectric layer of titanium dioxide (TiO2) and a monolayer of tungsten disulfide (WS2). When illuminated by the polarized light (x direction) with magnetic field $H$ in the y axis [Fig. 1(b)], the bare film-coupled nanocube in absence of the WS2 can support a strong PIMR mode by generating one pair of antiparallel currents in the nanocube and its mirror image in the silver film [31,32], which induce a diamagnetic response [29,33–36] with an electric current loop between real and imaging nanocube under the plasmon-hybridization effect [31,37]. The PIMR excitation can be readily confirmed by our simulation using finite difference time domain (FDTD) method. In our simulation, a total-field linearly polarized scattered-field (TFSF) source is employed to illuminate the nanocube. The thickness of monolayer WS2 is set to 1 nm. The side length of the silver nanocube is 23 nm–37 nm. To consider practically fabricated cube geometry and to avoid numerical artifacts, all the corners and edges of the nanocube were rounded with a rounding radius of 3 nm. The areas covering the nanocube are all set to a 0.5 nm grid. In order to avoid nonphysical reflections around the nanocube, we adopted perfect matching layer boundary conditions for the simulation domain. Figure 1(c) plots the simulation result of the scattering spectrum for the bare NPoM system with the cube length $L = 31$ nm and the film thickness of $t = 7$ nm. It can be clearly seen that a strong scattering peak occurs around 2.02 eV. The local electric [Fig. 1(d)] and magnetic [Fig. 1(e)] field distributions at the peak wavelength further confirm this resonance as the PIMR. The input oscillating magnetic component induces a circulating current loop around the nanocube and its mirror image [red arrows in Fig. 1(d)], thereby producing a diamagnetic response with a “magnetic dipole” $\mu_m$ orienting in the y direction. This dipole, in turn, is coupled to the film-coupled system to generate a strong magnetic resonance [31,33,34] with a strong localized magnetic field inside the TiO2 layer, as shown in Fig. 1(e). It is important to note that the corresponding electric field at the magnetic resonance shows much improved field confinement with unique feature in dipolar emission, which plays an important role in obtaining large coherent coupling energy with reduced incoherent coupling strength. We will discuss this point in a later section.

To investigate the strong coupling between the single nanocube and the WS2 monolayer, we vary the cube length to tune the magnetic mode to be on resonance with the
FIG. 2. (a) Scattering spectra (in color scale) of the hybrid system as a function of nanocube length overlapped with the dispersions of uncoupled (dashed white lines) and coupled (dashed black lines) modes. (b) Simulated (solid black) and fitted (dashed red) spectra of the hybrid system at zero detuning ($L = 31\text{ nm}$). The simulated (solid curves) and fitted (squared curves) dispersion relationship (c) and the spectral width (d) of the hybrid system.

Here, $a_{\phi}$ denotes the background amplitude, $\phi_{k}$ represents the spectral phase of the hybrid states and $b_{k}$ is the amplitude. A good match of the fitted spectra (dashed red line) to the simulated results (solid black line), as shown in Fig. 2(b), proves that the inhomogeneous broadening of the hybrid polarization modes may not exist or only possess a small contribution to their scattering spectra, which can be ignored in the present case.

Figures 2(c) and 2(d) give the extracted dispersion ($\omega_{k}$) and the spectral full width at half maximum (FWHM, denoted by $2\gamma_{k}$) of the coupling system, respectively. The fitted dispersion relationship (blue squares) of the hybrid system shows significant anticrossing behavior with a normal mode splitting up to $\hbar\Delta_{\text{SNSM}} = 2\hbar|\Omega_{S}| \approx 280 \text{ meV}$ at zero detuning $\Delta = |\omega_{p} - \omega_{X}| = 0 \text{ (L = 31 nm)}$. This is so far the largest value of Rabi splitting obtained in the strong coupling between the monolayer of TMDs and the single nanoparticle at ambient temperature. It is important to note that the spectral width of the UP branch exceeds the LP branch by 5 meV at zero detuning, which indicates the existence of incoherent coupling process via the continuum reservoir resulting in the formation of sub/superradiant polariton states [7,15,32,39].

The coherent and incoherent coupling process can be quantitatively described by the widely-used coupled oscillator model with a non-Hermitian Hamiltonian [14,40]:

$$\hat{H} \left[ \Omega_{S} \frac{\hat{\omega}_{X}}{\hat{\omega}_{p}} - i \left[ \frac{\gamma_{e}}{\gamma_{c}}, 0 \right] \right] \left[ \alpha, \beta \right] = E \left[ \alpha, \beta \right].$$

Here, $\tilde{\omega}_{X} = \omega_{X} - i\gamma_{X}$ and $\tilde{\omega}_{P} = \omega_{P} - i\gamma_{P}$ give the complex resonance frequencies of the uncoupled WS$_2$ excitonic mode and the PIMR mode, respectively. $\alpha$ and $\beta$ are eigenvector components, i.e., Hopfield coefficients, which satisfy $|\alpha|^2 + |\beta|^2 = 1$. The dispersions and damping rates of the polariton modes are given by the real and imaginary parts of the complex eigenvalues:

$$E_{k} = \hbar\tilde{\omega}_{k} = \hbar \left[ \frac{\tilde{\omega}_{X} + \tilde{\omega}_{P}}{2} \right] \pm \frac{1}{2} \sqrt{\left( \frac{\tilde{\omega}_{X} + \tilde{\omega}_{P}}{2} \right)^2 + \left( |\Omega_{S}|^2 - \gamma_{c}^2 \right) - 2i\gamma_{e} \text{Re}(\Omega_{S})}.$$  

By diagonalizing Eq. (3) with optimal coherent coupling strength $\hbar|\Omega_{S}| = 140 \text{ meV}$ and the incoherent coupling strength $\hbar\gamma_{e} = 3 \text{ meV}$, we are able to reproduce both the polariton energetics and spectral width of the polariton modes, as shown by the solid lines in Figs. 2(c) and 2(d), respectively. The coherent coupling constant is generally a complex quantity, which can be given as $\Omega_{S} = |\Omega_{S}| e^{i\phi_{S}}$ with the Rabi phase $\phi_{S}$ describing the spatial distance and the resultant relative phase difference between the two individual subsystems. Our previous work reveals the microscopic origin of the Rabi phase: It originates from the initial phase difference between two individual modes which cannot be eliminated by a U(1) gauge transform [41]. Here, a negative value is used for the coherent coupling constant, i.e., $\hbar\Omega_{S} = -140 \text{ meV}$, which indicates that the Rabi phase $\phi_{S} = \pi$. Apparently, the present hybrid system offers a greatly improved coherent coupling strength with a prominent reduction of incoherent coupling process with respect to those in our previously reported hybrid
FIG. 3. The normalized near-field distribution of the electric components at the surface right below the nanocube in the gap at the PIMR frequency for cube length of $L = 31$ nm and dielectric thickness of $t = 7$ nm.

NPoM system with single nanorod exhibiting much stronger incoherent coupling strength over 16 meV [32]. Note that both coherent and incoherent coupling can be described by a more formal model for full open quantum systems, as described in Refs. [39,42].

B. Coherent and incoherent coupling strength

To explore the underlying mechanism behind the enhanced coherent normal mode splitting with much reduced incoherent coupling strength, we quantitatively evaluated the near-field characteristics of the NPoM system. Figures 3(a)–3(c) demonstrate the near-field distribution of the electric components in the $x$-$y$ plane right below the nanocube at the resonance energy of the magnetic mode normalized to the input electric field $E_0$ for cube length of $L = 31$ nm and dielectric thickness of $t = 7$ nm. Several aspects can be clearly seen: (i) the electric field is mainly localized at either the edges ($x$ and $z$ component) or the corners ($y$ component) of the nanocube, (ii) the electric field in $z$ direction is enhanced at the vertical edges (parallel to the $y$ axis) of the nanocube with the maximum enhancement factor up to 52 [Fig. 3(c)], and importantly, (iii) considerable contribution to the field confinement can also be found from both the $x$ and $y$ component, which provides up to a maximum of 20-fold enhancement in the amplitude with respect to that of the incident electric field, as shown in Figs. 3(a) and 3(b).

Under the excitation of a plane wave impinging normally to the film surface, the nanocube can support the gap plasmon with the magnetic flux passing through the gap. Such a gap mode generally exhibits strong field confinement in the normal direction [43], which has been observed and demonstrated in other film-coupled nanocube systems [26,44]. In our case, the configuration of the coupled system is optimized (including the high-refractive-index dielectric material, the gap thickness, and the cube size) such that the in-plane ($x$ and $y$ components) intensities of the electric field at the magnetic resonance are comparable to that of the $z$ component. These features greatly facilitate the coupling of the in-plane $\text{WS}_2$ excitons to the local electric field, thus accounting for the present large normal mode Rabi splitting. Note that in the system of nanocube coupled to the $\text{WS}_2$ monolayer reported in Ref. [26], the maximum normalized field intensity (around $7 \sim 12$) in the $z$ direction is similar to our results, but the corresponding in-plane intensities are much weaker, which may be one of the reasons for the observed small coherent coupling energy.

Another key feature of this film-coupled system lies in the fact that the in-plane electric field at the magnetic resonance is confined not only at the cube edges but also in the ultrathin $\text{TiO}_2$ layer, which behaves as an open nanocavity and therefore offers a squeezed mode volume to enhance the strong light-matter coupling. For the case in Fig. 1(d) giving the electric field distribution in the $x$-$z$ plane for the thickness of $t = 7$ nm and cube length $L = 31$ nm, based on the expression $V = (f \cdot H^2 dV^2)/(f \cdot H^2 dV^2)$, the corresponding mode volume can be readily numerically obtained $V = 3000 \text{nm}^3$, which is smaller than the mode volume of other similar structures [26,28]. In this sense, we can further calculate the number of excitons $N$ that are involved in the strong coupling according to the relationship $g = \sqrt{N \mu_c |E|} = \sqrt{N \mu_c \sqrt{\hbar \omega/2 \epsilon_0 \delta V}}$ [22,28,32]. Here, $\omega$ and $\epsilon$ represent the angular frequency and the permittivity of $\text{TiO}_2$, respectively. Considering the Rabi splitting up to 280 meV and the transition dipole moment of an exciton $\mu_c \approx 50$ D in the $\text{WS}_2$ monolayer [7,22,32], the number of excitons ultimately involved in the strong coupling process can be estimated to be $N \approx 7$. In our previously reported nanorod-based hybrid system, we have demonstrated a similar number of excitons coupling to the magnetic mode but with much smaller Rabi splitting, which is attributed to the configuration of the nanorod system offering relatively larger mode volume with weaker in-plane field localization [32].

Despite the large coherent coupling energy, the hybrid system exhibits a surprisingly reduced incoherent coupling strength. Microscopically, incoherent coupling generally occurs as two strongly coupled subsystems exchange their energy by spontaneously emitting photons from one system first to the vacuum continuum reservoir; these photons are, in turn, reabsorbed by the other subsystem without conserving any phase relationship [39]. This process, as a coupling-induced phenomenon, provides an incoherent damping channel and can eventually modulate the radiative damping rates with the formation of sub- and super-radiant hybrid modes [7,15,32]. The incoherent coupling strength characterized by the coupling-induced cross-damping term $\gamma_c$ is given as [39]:

$$\gamma_c \leq \sqrt{\gamma_{\text{X,rad}} \gamma_{\text{Prad}}} (\mu_\text{X} \cdot \mu_\text{E})^2,$$

which is related to both the radiative damping rates ($\gamma_{\text{X,rad}}$ and $\gamma_{\text{Prad}}$) and the inner product of the unit vector of the dipole moments determined by the orientation of the dipole moments of the subsystems.

To give a deep insight on the drastic reduction in the incoherent coupling strength, we quantitatively studied the scattering property of the magnetic dipole by simulating the far-field emission pattern of the film-coupled nanocube at the dipolar resonance. Figure 4(aii) plots the radiation map (in color scale) in the $x$-$y$ plane for $t = 7$ nm with the radiation intensity normalized to unity. Apparently, the far-field radiation at the PIMR resonance exhibits good directivity in the $z$ direction, indicating that the electric dipole moment of the magnetic plasmon $\mu_\text{E}$ is mainly orientated normal to the film surface, as marked by the dashed white circle. The detailed emission feature can be further quantitatively demonstrated by the directivity cross section in the $x$-$z$ plane. As shown in Fig. 4(b), the radiation pattern of the film-coupled nanocube at the PIMR frequency exhibits...
a single lobe oriented in the cube surface (z direction) with its radiation falling mainly inside the orientation angle of $-40^\circ \sim 40^\circ$ (gray curve). This emission characteristics can reasonably explain the reduced incoherent coupling strength based on Eq. (5): The incoherent exchange of photons between the excitonic and plasmonic systems via the continuum reservoir can be effectively suppressed due to the effective dipole moment $\mu_p$ that is generally orientated normal to the in-plane excitonic dipole $\mu_X$, giving rise to a minimized amplitude of the inner product in Eq. (5). From this point, the mutual orientations between these two effective dipoles can be further quantitatively estimated by considering the radiative damping rates of the subsystems applied in the present case.

Suppose a high-quality exfoliated WS$_2$ flake (free-standing or deposited on a quartz substrate) at room temperature, in our simulation, we applied a typical linewidth for the excitonic resonance, i.e., $\hbar\Gamma_X = \hbar\Gamma_{X,\text{rad}} + \hbar\Gamma_{X,\text{nonrad}} = 50\,\text{meV}$, which is generally assumed to be homogeneously broadened with the contributions from both radiative ($\Gamma_{X,\text{rad}} = 2\gamma_{X,\text{rad}}$) and nonradiative ($\Gamma_{X,\text{nonrad}} = 2\gamma_{X,\text{nonrad}}$) broadening [7,45]. Note that here we neglect the broadening introduced by the pure dephasing processes [32]. Spectroscopic measurements have shown that the total spectral width is mainly governed by the nonradiative decay and the contribution of radiative decay is very small, which give a linewidth broadening of $\hbar\Gamma_{X,\text{rad}} \approx 7\,\text{meV}$ [46], corresponding to the radiative damping rate $\hbar\gamma_{X,\text{rad}} \approx 3.5\,\text{meV}$. For the PIMR resonance, radiative damping is expected to dominate the linewidth broadening with negligible contribution from nonradiative damping and pure dephasing process. At zero detuning, a fit of the scattering spectrum [Fig. 1(c)] to a Lorentzian lineshape gives the PIMR spectral width $\hbar\Gamma_{\text{PIMR}} \approx 2\hbar\gamma_{\text{PIMR}} \approx 190\,\text{meV}$. This results in the radiative damping rate $\hbar\gamma_{\text{PIMR}} \approx 95\,\text{meV}$. According to Eq. (5), we can therefore estimate the mutual orientation of these two effective dipole moments, which are nearly perpendicular to each other with an angle of $80^\circ$.

Such a unique far-field radiation aspect of the film-coupled nanocube with small radiative damping rate is advantageous for the study of coherent strong light-matter interactions compared with the traditional plasmonic system coupled to organic semiconductors studied in our previous work [15]. In that hybrid system, relatively stronger radiative damping rate of the ensemble of excitons in organic J-aggregate dye molecules and the effective plasmonic dipole mainly orientated within the planar gold film leads to a much stronger incoherent coupling strength.

Now we discuss how to effectively control both the coherent and incoherent coupling strength in the present hybrid system. Essentially, the coherent coupling strength is extremely sensitive to the thickness of the dielectric layer. In order to further explore the influence of the thickness of the dielectric layer on the coupling strength of the hybrid system, we first calculated dependence of Rabi splitting on the mode volume of the hybrid system for different thickness, as shown in Fig. 4(c). The mode volume increases dramatically as the dielectric TiO$_2$ layer becomes thicker. Accordingly, the Rabi splitting featuring the coherent coupling strength decreases down to 200 meV for the thicker layer with $t = 20\,\text{nm}$. This is expected because of the weakening of the confined electric field in the dielectric layer [28]. In principle, the Rabi splitting can be further improved by shrinking down the film thickness, however, ultrathin layer, for example, 3- or 4-nm-thick TiO$_2$ in the present film-coupled nanocube system will cause a considerable redshift of the PIMR mode (results are not shown here). In this sense, we need to decrease the cube size down to sub-10 nm to make the PIMR mode resonant with the excitonic resonance at $\hbar\omega_X \approx 2.02\,\text{eV}$, which makes the practical fabrication very challenging. Moreover, decreasing the thickness of the dielectric film will also generate other electric dipole modes occurring very close to the PIMR modes. These plasmonic modes can also strongly couple to both the PIMR mode and the excitonic resonance at zero detuning, resulting in much more complicated light-matter hybridization.

Interestingly, we find that the far-field emission pattern of the PIMR mode can be readily controlled when adjusting the film thickness as well, which can further influence the incoherent coupling strength. We demonstrated the calculated radiation patterns in Figs. 4(ai)–4(aiii) for different film thickness. When the film thickness increases up to 14 nm, the emission directivity is improved, as marked by the smaller dashed red circle in Fig. 4(aii) with respect to the dashed white circle in Fig. 4(ai). The dipolar resonance radiates tightly with a single lobe falling mainly inside the orientation angle of $-30^\circ \sim 30^\circ$ (red curve in Fig. 4(b)) in the z direction. The directivity pattern does not change so much as the film thickness keeps increasing over 20 nm, as demonstrated by the blue and red curve in Fig. 4(b). Note that the spectral width of the PIMR...
mode at the excitonic resonance almost keeps constant for different film thickness, as shown in Fig. 4(d). This indicates that the radiative damping rates \((γ_{\text{P, rad}}})\) of the plasmonic system does not change as a function of thickness \(t\). Therefore, according to Eq. (5), one can readily reduce the incoherent coupling strength by employing thicker dielectric layer under the nanocube. This means that coherent/incoherent coupling strength is enhanced/suppressed simultaneously with the decrease/increase of the film thickness. In the present case, we have carefully optimized the structure configuration such that a large Rabi splitting can be obtained with an appropriately suppressed incoherent coupling strength.

### III. CONCLUSIONS

In conclusion, we demonstrate the strong coupling between the magnetic plasmon generated by the film-coupled nanocube system and the A exciton in the monolayer WS\(_2\) through a combination of numerical simulation and theoretical analysis. With the help of plasmon-induced magnetic resonance induced in the film-coupled nanocube, we obtained an enhanced coherent PIMR-exciton coupling over 280 meV due to the much higher in-plane field confinement at the cube edges and inside the dielectric cavity at room temperature. We further analyzed the underlying mechanism for the suppressed incoherent coupling strength, which is mainly attributed to the small radiative damping rate of the excitonic resonance and the net electric dipole moment of the PIMR orientating perpendicular to the film surface. The numerical simulations and theoretical analysis reveal that coherent/incoherent coupling strength can be simultaneously enhanced/suppressed with the decrease/increase of the film thickness. This is expected to be useful for controlling the coherent and incoherent coupling process in strongly coupled TMD-based NPoM systems, which makes such hybrid nanostructure a good platform for the study of strong light-matter interaction and for the potential applications in realizing novel quantum and nanophotonic optical devices.

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