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Dark state physics has led to a variety of remarkable phenomena in atomic physics, quantum optics, and information theory. Here, we investigate interacting dark resonance type physics in multi-layered plasmonic meta-molecules. We theoretically demonstrate that these plasmonic meta-molecules exhibit sub-natural spectral response, analogous to conventional atomic four-level configuration, by manipulating the evanescent coupling between the bright and dark elements (plasmonic atoms). Using cascaded coupling, we show nearly 4-fold reduction in linewidth of the hybridized resonance compared to a resonantly excited single bright plasmonic atom with same absorbance. In addition, we engineered the geometry of the meta-molecules to realize efficient intra-molecular excitation transfer with nearly 80%, on resonant excitation, of the total absorption being localized at the second dark plasmonic atom. An analytical description of the spectral response of the structure is presented with full electrodynamics simulations to corroborate our results. Such multilayered meta-molecules can bring a new dimension to higher quality factor plasmonic resonance, efficient excitation transfer, wavelength demultiplexing, and enhanced non-linearity at nanoscale. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4896035]

Quantum emitters (atoms, molecules, and quantum dots) prepared in a specific coherent superposition of states exhibit many remarkable phenomena in atom optics,[1] cavity quantum electrodynamics (cQED),[2] quantum computation, and quantum information.[3] For instance, dark state in three-level atoms in (Λ)-configuration holds the key for many counter-intuitive effects in atomic physics and quantum optics.[4] Recently, the domain of dark resonances has extended beyond its conventional plasmonic atomic, molecular, and optical physics to plasmonics and metamaterials. Several classical or plasmonic analogue of quantum optical phenomena like plasmon induced transparency (PIT),[5,6] electromagnetic induced absorption (EIA),[7,8] have created new interests in dark state physics with applications such as sensing,[9] broadband slow light,[10] ultra-sensitive spectroscopy,[11] and plasmonic ruler.[12,13]

However, extremely poor (metal-loss-limited) quality factor of the plasmonic resonance often pose a serious bottleneck for practical applications. Incorporating gain medium[14] to compensate the loss is one way to go which has its own challenges.[15,16] Recently giving a quantum boost to plasmonic devices[17,18] has been proposed to make an efficient use of the phase coherent gain medium coupled to such systems.

In this letter, we theoretically investigate a multi-layered plasmonic meta-molecule consisting of a radiative (bright) and cascaded subradiant (dark) elements, as shown in Fig. 1(c), to realize interacting dark resonances type physics. We show that these meta-molecules exhibit sub-natural spectral response analogous to atomic four-level configuration.[19] A 4-fold reduction in linewidth of the hybridized resonance is achieved, compared to resonantly excited single bright

plasmonic atom with same absorbance. These engineered meta-molecules also exhibit efficient intramolecular excitation transfer and ∼80% (of the total absorption) is localized at the second dark element, on resonant excitation. On the other hand at off-resonant excitation, ∼72% is localized at the first dark plasmonic atom. Such wavelength dependent localization of excitation may open new possibilities for wavelength demultiplexing at nanoscale.

One generic four-level model displaying double-dark resonance (DDR), in conventional atomic physics, is shown in Fig. 1(a). The dark-state of the Λ-configuration is coupled to the metastable state $|d\rangle$ by the control field. The existence of DDR is apparent in the presence of two different Λ-subsystems, as seen in Fig. 1(b). Within the transparency window, of the Λ-configuration atomic electromagnetic induced transparency (EIT), the interference induced by coherently interacting dark-states leads to sharp resonances.[19,20] Geometry of the multilayered meta-molecule displaying interacting dark resonance type physics is shown in Fig. 1(c). Here, we used the dipolar mode of a gold bar ($l_1$nm × w nm × t nm) and the subradiant asymmetric mode of two parallel gold bars ($l_2$nm × w nm × t nm) to serve as the bright and dark plasmonic atom, respectively. Energy level diagram of the meta-molecule is shown in Fig. 1(d) where the coupling between the bright-dark and dark-dark plasmonic atoms is via near-field interaction. The incident electromagnetic field $E = \mathcal{E}e^{-i\omega t}$ couples strongly with the radiative plasmonic state $|a_0\rangle = \tilde{a}_0(\omega)e^{-i\omega t}$ and weakly with the dark plasmonic states $|a_\alpha\rangle = \tilde{a}_\alpha(\omega)e^{-i\omega t}$ (where $\alpha = 1$, 2). The decay of the dark plasmonic atoms $\gamma_{1,2}$ (ohmic losses, weak coupling to radiation, and intrinsic dipole moments) is much weaker than the bright plasmonic atom γ_0 where the loss is dominated by radiation damping.[21] The
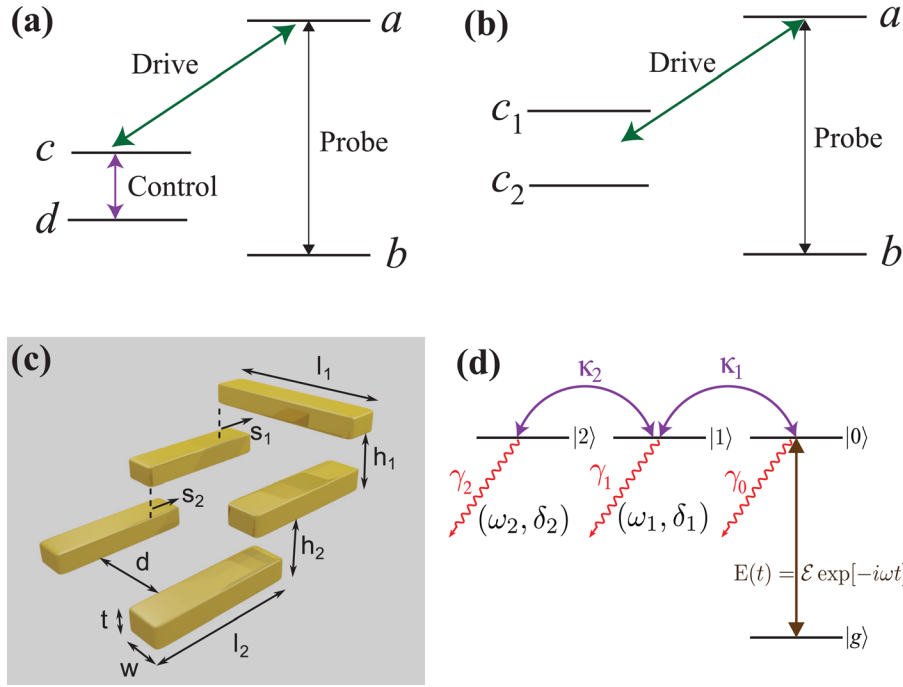
FIG. 1. (a) Atomic four-level system displaying double-dark resonance. (b) Four-level system obtained by diagonalizing the interaction with the control field. Here, the drive field can couple both dressed states $|c_{1,2}\rangle$ with state $|a\rangle$ thus forming two Λ subsystems with interacting dark resonances. (c) Isometric view of the multi-layered stacked geometry of the plasmonic meta-molecule. (d) Equivalent energy level diagram of the plasmonic meta-molecule with near-field couplings $\kappa_{1,2}$. Here, we have defined the detuning as $\delta_\alpha = \omega_0 - \omega_\alpha$ with $\alpha = 1, 2$.

coupling between the bright-dark and dark-dark plasmonic atoms is given by the parameters κ_1 and κ_2, respectively. The amplitudes of the three states can be obtained by the linearly coupled Lorentzian oscillators (LCLOs) model.[23]

Solving for the dipolar response of the radiative plasmonic atom we get

$$\chi \sim \frac{ig\Gamma_1}{\Gamma_1\Gamma_0 + |\kappa_1|^2}\left\{1 + \frac{|\kappa_1|^2|\kappa_2|^2}{\Gamma_1\left[\Gamma_0\left(|\kappa_2|^2 + \Gamma_1\Gamma_2\right) + |\kappa_1|^2\Gamma_2\right]}\right\}, \tag{1}$$

where we have defined the damping $\Gamma_\alpha = \gamma_\alpha + i(\omega - \omega_\alpha)$, ($\alpha = 0, 1, 2$) and the parameter g quantifies the strength of the coupling between the bright plasmonic atom with the incident electromagnetic wave. The functional form of χ reminds us of the linear susceptibility for an equivalent atomic media where the coherent drive field $\Omega_{1,2}$ control the dynamics of the dark resonance.[19] In the plasmonic analogue, the role of $\Omega_{1,2}$ is played by the near field couplings $\kappa_{1,2}$. Due to the second term in Eq. (1), an absorptive resonance emerges in the spectral response of the bright atom. We can approximate the expression for $\tilde{a}_0$ (obtained from the LCLO model) around $\omega \sim \omega_1$ assuming $|\kappa_2| \ll |\kappa_1|$, and $\omega_0 = \omega_1 = \omega_2$ (to keep the final expression simple) as

$$\tilde{a}_c = \frac{|\kappa_2|^2}{|\kappa_1|^2}\left[\frac{g\mathcal{E}}{\Delta - i\gamma_0\left(\gamma_2/\gamma_0 + |\kappa_2|^2/|\kappa_1|^2\right)}\right]. \tag{2}$$

Equation (2) has a typical Lorentzian form with linewidth (full width at half-maximum)

$$\tilde{\gamma}_c = 2\gamma_0(\gamma_2/\gamma_0 + |\kappa_2|^2/|\kappa_1|^2), \tag{3}$$

which in the limit of $\gamma_2 \to 0$ reduces to $2\gamma_0(|\kappa_2|^2/|\kappa_1|^2)$. If we compare Eq. (3) with the dipolar response of the bright plasmonic atom in the absence of any dark plasmonic atoms,

the amplitude and the linewidth are scaled as $|\kappa_2|^2/|\kappa_1|^2$ and $(\gamma_2/\gamma_0 + |\kappa_2|^2/|\kappa_1|^2)$, respectively.

Next, we present the numerical simulation to investigate the spectral response of the three-layered meta-molecule shown in Fig. 1(c). The dimensions of the bars were carefully tuned to match the resonance frequency of the bright and dark plasmonic atoms. The asymmetric position of the top layer gold bar with respect to the middle layer provides stronger near field coupling strength.[6] The bottom layer is identical to the middle in geometry, and they are partially overlapped. The gap between top and middle layers is $g_1 = (h_1 - t)$ nm and the gap between parallel gold bars is $d = 200$ nm. The gap between the lower two layers is $g_2 = (h_2 - 40)$ nm. The size of the unit cell was chosen so that the diffraction orders from the periodic structure do not interfere with the resonances of interest. We applied periodic boundary condition to simulate an array of such nanostructures. The dielectric constant for the gold is taken from Johnson and Christy[22] while the surrounding dielectric medium[23] is vacuum with $\epsilon = 1$.

In Fig. 2(a), we have plotted the absorbance of an isolated bright plasmonic atom (dashed blue), bright plasmonic atom coupled to a dark plasmonic atom also know as PIT structure (dotted dashed black), and multilayered meta-molecule with bright plasmonic atom coupled with two dark plasmonic atoms (solid red). The isolated bright plasmonic atom (355 nm × 80 nm × 40 nm) has a dipolar resonance at $\omega_0 \sim 264$ THz, and the spectral response linewidth (fullwidth at half maximum, FWHM) is $\tilde{\gamma}_1 = 2\gamma_1 \sim 38.1$ THz which is dominated by the radiative damping. In the presence of a dark plasmonic atom, the structure exhibits EIT-type phenomena as seen by a reduction in absorbance at $\omega \sim 264$ THz. The basic physical mechanism behind this EIT-type phenomena can be understood as the destructive interference between the direct excitation of the radiative plasmonic atom and the back-action from the dark to the radiative plasmonic atom.[5] However, in the presence of a second dark plasmonic atom, the EIT-type transparency at
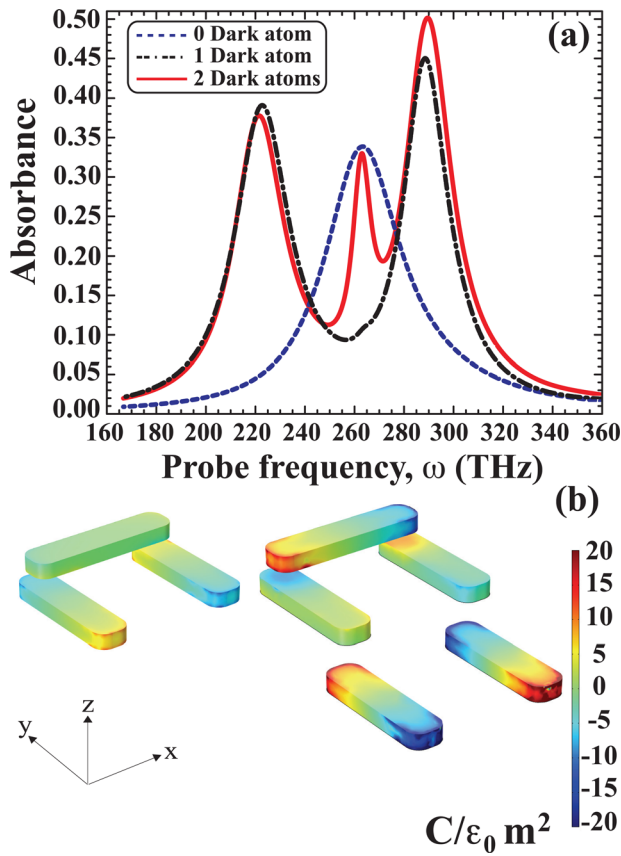
FIG. 2. (a) Plot of the absorbance of an isolated bright plasmonic atom (dashed blue), bright plasmonic atom coupled to a dark plasmonic atom (also know as PIT structure) (dotted dashed black), and multilayered meta-molecule with a bright plasmonic atom (cascaded) coupled with two dark plasmonic atoms (solid red). The central resonance, with absorbance equal to that of a resonantly excited bright plasmonic atom, has a linewidth (FWHM) which is more than 4-fold narrower. (b) Plot of the charge distribution along the bright and dark plasmonic atoms for probe excitation at $\omega = \omega_1 \sim 264$ THz. For the two dark plasmonic atoms geometry, the gaps $g_{1,2}$ are 30 nm and 210 nm, respectively, and the other parameters are $l_1 = 355$ nm, $l_2 = 315$ nm, $w = 80$ nm, $h_1 = 70$ nm, $s_1 = 87.5$ nm, $s_2 = 82.5$ nm, and $t = 40$ nm.

$\omega = \omega_1$ splits into two creating new windows at $\omega_{\pm} \sim \frac{1}{2}$ $\left(\omega_1 + \omega_2 \pm \sqrt{4|\kappa_2|^2 + (\omega_1 - \omega_2)^2} \right)$ in the limit of $\gamma_{1,2} \sim 0$. These transparency windows are created due to destructive interference between the paths $|g\rangle \rightarrow |0\rangle$ and $|g\rangle \rightarrow |0\rangle \rightarrow |1\rangle \rightarrow |0\rangle$, and between the paths $|g\rangle \rightarrow |0\rangle$ and $|g\rangle \rightarrow |0\rangle \rightarrow |1\rangle \rightarrow |2\rangle \rightarrow |1\rangle \rightarrow |0\rangle$. We also observe the appearance of a new resonance which is superimposed on the transparency window of the PIT structure. Peak absorbance of the new absorptive resonance is equal to that of an isolated bright plasmonic atom and a linewidth which is more than 4-fold narrower. In the supplementary material, we have included the effect of substrate and show that linewidth narrowing is indeed robust. The gap between the bright and the first dark plasmonic atom is $g_1 = 30$ nm, while the gap between the two dark plasmonic atoms is $g_2 = 210$ nm. For these gaps, the coupling strengths $\kappa_{1,2}$ are ~ 38 THz and ~ 8 THz, respectively. In atomic physics, the presence of such an ultra-sharp resonance is a signature of quantum interference.[19] In Fig. 2(b), we have plotted the charge distribution on all the elements at resonant excitation for the PIT and DDR structure with $g_1 = 30$ nm and

$g_2 = 210$ nm. We see significant charge distribution on the second dark plasmonic atom with respect to the first in DDR configuration which opens the possibility for efficient intra-molecular excitation transfer via coupled dark plasmonic atoms.

In Fig. 3(a), we have plotted the linewidth and the amplitude of the central resonance (absorbance curve $\omega = \omega_1 \sim 264$ THz) against the gap g_2 between the dark plasmonic atoms. To extract the linewidth information of the central resonance, we fitted the simulation data with three Lorentzian. We see that the linewidth decreases (monotonically) with g_2 and for large gap it approaches to the drude-damping limit value of $\tilde{\gamma} \sim 6.5$ THz for bulk gold.[22,24] However, the amplitude of the resonance has a interesting non-monotonic behavior. At first, it increases (monotonically) with the gap and attains the maximum value of ~ 0.48 at $g_2 \sim 130$ nm and begins to decrease thereafter. To quantify the quality factor of the new resonance, let us define the figure of merit as

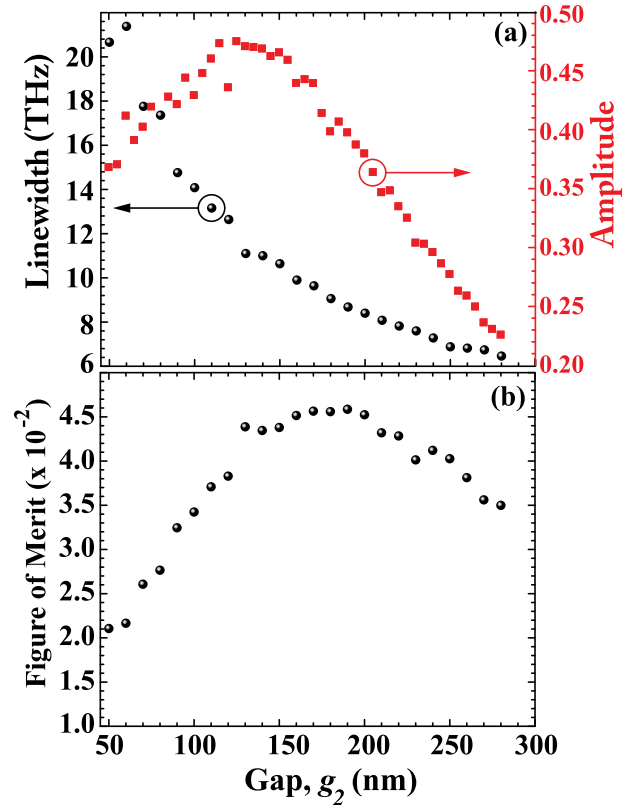$$\text{FOM} = \frac{\text{Absorbance}(Amplitude)}{\text{Linewidth}(FWHM)}. \tag{4}$$

FIG. 3. (a) Plot of the linewidth and the absorbance (amplitude) of the central absorptive resonance $\omega = \omega_1 \sim 265$ THz against the gap between the dark plasmonic atoms. For weak dark-dark atom coupling (i.e., larger gap), the linewidth approaches to the drude damping limit value of $\tilde{\gamma} \sim 6.5$ THz (for bulk gold) and the amplitude is of the same order as resonantly excited bright plasmonic atom alone. The amplitude has a non-monotonic dependence on g_2 which attains the maximum value of ~ 0.48 when the gap $g_2 \sim 130$ nm. (b) Plot of the FOM for the plasmonic meta-molecule against the gap between the dark plasmonic atoms. Near the optimum gap $g_2 \sim 170$ nm, the FOM reaches $\sim 4.50 \times 10^{-2}$ which is nearly 5-fold higher compared to an isolated bright plasmonic atom value of $\sim 9 \times 10^{-3}$. Other geometrical parameters for numerical simulations are same as Fig. 2.

In Fig. 3(b), we have plotted the figure of merit (FOM) of the meta-molecule against the gap g_2 while keeping $g_1 = 30$ nm. For an isolated bright plasmonic atom, the FOM is $\sim 9 \times 10^{-3}$. By incorporating a second dark plasmonic atom and tuning the coupling κ_2, the FOM reaches a maximum value of 4.5×10^{-2} (at $g_2 \sim 170$ nm) which is 5-fold higher compared an isolated bright plasmonic atom. Such an enhancement in the quality factor of the absorbance opens the possibility of enhancing the non-linear response of the meta-molecule.

Next, we discuss the possibility of efficient intramolecular excitation transfer by engineering the geometry of our meta-molecule. To quantify the efficiency of transfer, we have defined η as the ratio of the absorption by the second dark plasmonic atom to the total absorption given by

$$\eta = \frac{|\tilde{a}_2|^2}{\sum_{\text{All plasmonic atoms}} |\tilde{a}_i|^2}. \tag{5}$$

Using the LCLO model, we obtain the efficiency η as[23]

$$\eta = \frac{(\kappa_1 \kappa_2)^2}{(\gamma_1 \gamma_2 + \kappa_2^2)^2 + (\gamma_2 \kappa_1)^2 + (\kappa_1 \kappa_2)^2}. \tag{6}$$

In Fig. 4(a), we have plotted the efficiency (η) as a function of the gap g_2 while keeping g_1 fixed at 30 nm. We see the bell shaped curved maximum around 170 nm at which the efficiency reaches $\sim 80\%$. However, this gap does not correspond to maximum absorption by the molecule, which happens at $g_2 \sim 130$ nm where the total normalized absorption is $\sim 48\%$ (as seen from Fig. 3(a)). The numerical simulation data are in excellent agreement with analytical calculation (solid red curve) using the LCLO model. In Fig. 4(b), we have plotted the absorbance by the individual elements (and the total) of the meta-molecule when the gap $g_2 = 170$ nm. The absorbance (at resonant excitation $\omega = \omega_1 \sim 264$ THz) is dominated by the second dark plasmonic atom (80%), which shows that most of the energy absorbed by the meta-molecule is localized at the second dark plasmonic atom, thus exhibiting efficient transfer of energy within the elements. If we move to red-detuned probe frequency at $\omega \sim 242$ THz, nearly 72% of the total absorption is localized at the first dark plasmonic atom, while the rest is shared equally among the bright and the second dark atoms. However, if we move to blue-detuned transparency window at $\omega \sim 274$ THz, the entire absorption is localized in the vicinity of the dark plasmonic atoms only. Such frequency response of the meta-molecule could be helpful for routing local fields via different plasmonic atoms.

In summary, we have investigated interacting dark resonance type physics with multi-layered plasmonic meta-molecules. Analogous to the conventional atomic physics, these meta-molecules exhibit sub-natural spectral response near resonant excitation. Using cascaded coupling, we show nearly 4-fold reduction in the linewidth of the hybridized resonance compared to a resonantly excited single bright plasmonic atom with same absorbance. We also theoretically demonstrate ~ 5-fold enhancement in the FOM. The geometry of meta-molecule can be engineered to demonstrate efficient transfer of excitation from the bright to the dark plasmonic atoms at both resonant and off-resonant excitation. On resonant excitation, we demonstrate $\sim 80\%$ of the total absorption to be localized at the second dark plasmonic atom. Recent development in the field of nano-fabrication will enable the demonstration of such designs for experimental realization. Such engineered multi-layered meta-molecules can bring a new dimension to efficient excitation transfer, wavelength demultiplexing, etc., at nanoscale regime.
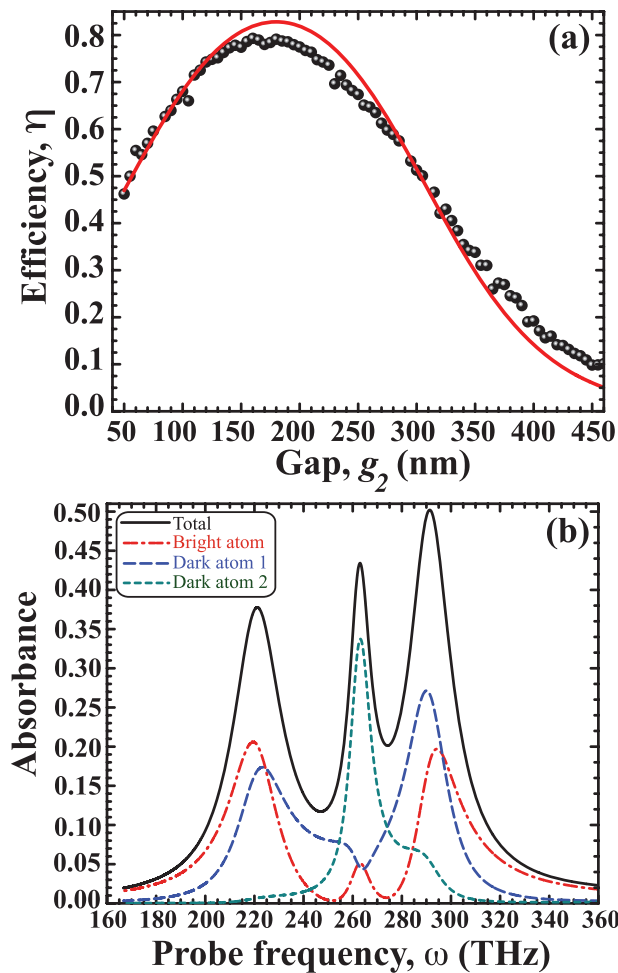
FIG. 4. (a) Plot of the efficiency (η) of the excitation transferred to the second dark plasmonic atom against the gap (g_2) between the two dark plasmonic atoms for resonantly ($\omega = \omega_1 \sim 264$ THz) excited meta-molecule. At the optimum gap of 170 nm, the efficiency is ~ 0.8. The solid red line (Eq. (6)) is in excellent agreement with the simulation data (black solid spheres). (b) Plot of the absorbance (total and individual contributions) when the gap between the dark plasmonic atoms at the optimum gap $g_2 = 170$ nm. The bright and the first dark plasmonic atom contributes equally to the absorbance of the meta-molecule which adds up to 20% and the rest 80% from second the second dark plasmonic atom. Other geometrical parameters for numerical simulations are same as Fig. 2.

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