Research article

Oscar Ávalos-Ovando*, Lucas V. Besteiro, Zhiming Wang* and Alexander O. Govorov*

Temporal plasmonics: Fano and Rabi regimes in the time domain in metal nanostructures

<https://doi.org/10.1515/nanoph-2020-0229>

Received April 6, 2020; accepted June 26, 2020; published online July 20, 2020

Abstract: The Fano and Rabi models represent remarkably common effects in optics. Here we study the coherent time dynamics of plasmonic systems exhibiting Fano and Rabi spectral responses. We demonstrate that these systems show fundamentally different dynamics. A plasmonic system with a Fano resonance displays at most one temporal beat under pulsed excitation, whereas a plasmonic system in the Rabi-like regime may have any number of beats. Remarkably, the Fano-like systems show time dynamics with very characteristic coherent tails despite the strong decoherence that is intrinsic for such systems. The coherent Fano and Rabi dynamics that we predicted can be observed in plasmonic nanocrystal dimers in time-resolved experiments. Our study demonstrates that such coherent temporal plasmonics includes non-trivial and characteristic relaxation behaviors and presents an interesting direction to develop with further research.

Keywords: Fano effect; plasmonics; time dynamics.

1 Introduction

The Fano and Rabi effects are characteristic properties of two interacting oscillators. For a system to exhibit the Fano effect (FE), one oscillator should have a narrow absorption resonance and the other one should be strongly damped [1]. In striking contrast to the Fano system, the two oscillators involved in a Rabi resonance have narrow absorption lines. Importantly, in both models, the involved oscillators should interact, and this interaction leads to interesting consequences. In the Fano system, the optical resonance possesses a peculiar line-shape, whereas the Rabi system exhibits a very characteristic splitting in its optical spectrum.

Modern nanostructures offer a variety of optical systems where the FE can be realized [2, 3]. In the field of nanostructures, prominent examples of FE come from exciton–plasmon interaction, plasmon–plasmon coupling, and the interaction of a localized exciton with a continuous spectrum [4–18]. Typically, the FE is observed in the spectral responses, such as absorption or scattering, which are optical measurements in the frequency domain. In contrast, this article focuses on manifestations of the FE in the time domain under pulsed excitations. We will contrast the time-resolved FE with the case of Rabi resonance in the time domain.

Regarding the Rabi regime, this effect has been observed in many experimental systems in both spectral and time-resolved domains. In hybrid exciton–plasmon nanostructures and in electromagnetic resonators with a built-in two-level system, this phenomenon is detected as the so-called Rabi splitting in the frequency domain [19–22]. For plasmonic nanocrystals, this spectral splitting effect is often regarded as a plasmon–plasmon hybridization [23]. In quantum semiconductor systems, it can be seen as the so-called Rabi oscillations in the time domain, when a two-level excitonic system interacts with a monochromatic electromagnetic wave [24–26]. In all the above cases, the Rabi-like regime involves two different excitations, both having narrow absorption lines.

This study investigates the Fano and Rabi regimes in the time domain using coupled plasmonic nanoparticles (NPs) as a model system. We found that the FE in the time domain exhibits very characteristic and unique features.

***Corresponding authors:** Oscar Ávalos-Ovando, Institute of Fundamental and Frontier Sciences, University of Electronic Science and Technology of China, Chengdu 610054, China; Department of Physics and Astronomy, Ohio University, Athens, OH 45701, USA; **Zhiming Wang**, Institute of Fundamental and Frontier Sciences, University of Electronic Science and Technology of China, Chengdu 610054, China; and **Alexander O. Govorov**, Institute of Fundamental and Frontier Sciences, University of Electronic Science and Technology of China, Chengdu 610054, China; Department of Physics and Astronomy, Ohio University, Athens, OH 45701, USA, E-mail: oa237913@ohio.edu (O. Ávalos-Ovando), jmwahng@gmail.com (Z. Wang), govorov@ohio.edu (A. O. Govorov). <https://orcid.org/0000-0003-3572-7675> (O. Ávalos-Ovando), <https://orcid.org/0000-0003-1316-6758> (A. O. Govorov)
Lucas V. Besteiro: Institute of Fundamental and Frontier Sciences, University of Electronic Science and Technology of China, Chengdu 610054, China; Centre Énergie Matériaux et Télécommunications, Institut National de la Recherche Scientifique, 1650 Boul. Lionel Boulet, Varennes, QC, Canada J3X 1S2, E-mail: lvbesteiro@gmail.com. <https://orcid.org/0000-0001-7356-7719>

The dynamical response of a Fano system is qualitatively different from that of a Rabi resonance. In the case of the Fano resonance, the coherent relaxation exhibits at most one beat in the dynamics and involves two relaxation exponents, fast and slow. In the Rabi-like systems, the relaxation dynamics may have a large number of beating oscillations before they are fully dampened. To reveal these features of coherent relaxation, we used two different excitation methods: point dipoles placed next to the coupled NPs and external electromagnetic pulses. The models used in our study include NPs made of both Drude and real noble metals. Our study aims to motivate a novel direction of research termed by us as “coherent temporal plasmonics,” in which custom-made plasmonic nanostructures could provide a very convenient platform for observing complex and non-trivial dynamical regimes.

2 Formalism

The formalism used in this study is based on the Maxwell's equations in the time domain and the local dielectric function model. We use Comsol Multiphysics software that specializes in the frequency-domain calculations, and, therefore, it will be convenient for us to utilize the Fourier transform. The displacement field, $\mathbf{D}$, in our formalism is given by (see Supporting Information)

$$\mathbf{D}(\mathbf{r}, t) = \int_{-\infty}^{\infty} e^{+i\omega t} \mathbf{D}_{\omega}(\mathbf{r}) d\omega = \int_{-\infty}^{\infty} e^{+i\omega t} \epsilon_0 \epsilon_{\omega}(\mathbf{r}) \mathbf{E}_{\omega}(\mathbf{r}) d\omega, \quad (1)$$

where $\mathbf{D}_{\omega}(\mathbf{r})$ and $\mathbf{E}_{\omega}(\mathbf{r})$ are the Fourier transforms of the displacement and electric fields, respectively; $\epsilon_{\omega}(\mathbf{r})$ is the local dielectric constant in the frequency domain and it should obey the property: $\text{Im } \epsilon_{\omega} < 0$ for $\omega > 0$. More details for this formalism are given in the Supporting Information.

Figure 1 shows the typical physical settings used in our study. To probe the coherent dynamics of coupled NPs, we either place an excitation dipole next to the NP pair (Figure 1a) or use an incident electromagnetic plane wave (Figure 1d). Assuming a pulsed excitation, the temporal responses of our system can be conveniently written via the corresponding Green functions, $G_E(\omega)$ and $G_d(\omega)$:

$$\begin{aligned} d_{\text{NC,E}}(t) &= \int_{-\infty}^{\infty} e^{+i\omega t} E_0(\omega) G_E(\omega) d\omega, \\ d_{\text{NC,d}}(t) &= \int_{-\infty}^{\infty} e^{+i\omega t} d_0(\omega) G_d(\omega) d\omega, \end{aligned} \quad (2)$$

where $E_0(\omega)$ and $d_0(\omega)$ are the Fourier transforms from the external electric field (x -component) and the exciting dipole, respectively. Then, the total dipole moment in an

NP dimer is defined as $d_{\text{tot}} = d_{\text{NP1}} + d_{\text{NP2}}$. For both types of excitation, we used short Gaussian pulses. In the case of the exciting dipole, the pulse function is given by $d_0(t) = d_0 \exp[i\omega_0 t - (t/\Delta t)^2]$, where d_0 , ω_0 , and Δt are the pulse parameters: the amplitude, the central frequency, and the pulse duration, respectively.

3 Results

We now begin constructing our main model systems (Rabi- and Fano-like) by incorporating NPs made of “Drude” metals (Figure 1). These cases allow us to understand general properties of the Fano and Rabi dynamics without dealing with the complications of real metals, such as interband transitions. Therefore, we take the dielectric constant of our NPs in the standard form (see Supporting Information and [27]):

$$\epsilon_i = \epsilon_{i,b} - \frac{\omega_{i,p}^2}{\omega(\omega - i\gamma_i)}, \quad (3)$$

where i is the index running over the NP labels. The medium outside the NPs is water (i.e., $\epsilon_w = 1.8$). We model two Drude dimers involving two dielectric functions, Drude₁ and Drude₂, which define the sharp and broad resonances, respectively (Figure 1). The corresponding Drude parameters can be found in Supporting Information. The Rabi dimer includes two NPs with the same dielectric constant Drude₁. The Fano dimer in Figure 1d comprises two nanorods with different dielectric functions. Although these cases are only a physical illustration, we found that the results obtained for the time dynamics are very generic. For the Rabi dimer, we first calculated the extinction cross section for one component (Figure 1b, left), and we observed only one sharp dipolar resonance at 481 nm. Then, when the dimer system is excited with a point dipole pulse $d_0(t)$, we see that the absorption spectra exhibit multiple peaks. The two main resonances around 475 and 508 nm (Figure 1b, right) correspond to the strongest anti-symmetric and symmetric modes, denoted as A- and S-modes (Figure S2). The time dynamics for the total moment is shown in Figure 1c. This trace shows a sustained oscillation with time, with several beats and nearly no decay in the time range shown, long after the initial pulse duration (shown in blue). Longer times start to show the decay. The frequency of the beats corresponds well to the difference between the frequencies of the main modes in the dimer, $\omega_{\text{A-mode}} - \omega_{\text{S-mode}}$, in Figure 1b.

Regarding the Fano dimer, NP₂ shows a broad resonance in the extinction owing to its dielectric constant, Drude₂, while NP₁ exhibits a sharp peak (Figure 1e). In the

Figure 1: Schematic and properties of Drude–Rabi (a,b,c) and Drude–Fano (d,e,f) systems. (a) Energy-level scheme (left) and the geometry of the Rabi dimer under dipole excitation (right). (b) Extinction of one isolated NP (left) and the absorption spectrum of the dimer (right). (c) Dimer’s time dynamics of the total dipole moment, including the excitation pulse (blue). Similarly, in the following panels, we show the optical properties of the Fano dimer. (d) Energy diagram of the Fano dimer. (e) Extinctions of isolated NPs. (f) Dimer’s extinctions and time dynamics. For the Rabi system, we used $R_{NP1} = R_{NP2} = 5 \text{ nm}$, gap = 1 nm, $\Delta t = 5 \text{ fs}$, $\lambda_0 = 495 \text{ nm}$. For the Fano system, we took $R_{NP1} = 1 \text{ nm}$, $L_{NP1} = 6 \text{ nm}$, $R_{NP2} = 5 \text{ nm}$, $L_{NP2} = 30 \text{ nm}$, gap = 1 nm, $\Delta t = 3 \text{ fs}$, and $\lambda_0 = 608 \text{ nm}$, where $\lambda_0 = 2\pi c/\omega_0$. For the linearly polarized light, we assume a wave polarized along z , incoming with $k||x$.

dimer configuration, these two, very different resonances will interfere destructively and give rise to a Fano resonance lineshape in the extinction, as shown in Figure 1f

(and Figure S1b). Let us now look at the time evolution of the Fano dimer upon excitation with a 3-fs electromagnetic pulse. We observe now an interestingly different response:

no beats and a sustained oscillation of the dimer's total dipole moment at long times. The slow exponential decay for the signal's envelope with an exponent of $\tau_s \sim 45$ fs persists for long after the initial pulse has ended, as shown in Figure 1f. This slow exponent corresponds to the damping of an excitation that is mostly localized in NP₁. However, NP₁ in our dimer is strongly interacting with NP₂, and the exponent τ_s is significantly reduced from the original decay in the NP₁, $2/\gamma_{NP_1}$. At short times just after the pulse, we can also see another, faster relaxation, with an exponent of $\tau_f \sim 3$ fs. This exponent comes from the fast decay inside the strongly dissipative NP₂. The single maximum in the amplitude comes from the confluence of two reasons: the initial pulse and the fast decay dynamics in NP₂. As we will soon see with more examples below, the whole pattern of relaxation in Figure 1f is very characteristic for the physical Fano system.

The above model systems provided us with interesting physical behaviors, and now it is time to look at these two regimes in real materials. To show that the properties seen in the Drude models are general, we now turn to model realistic materials such as gold and silver for the NPs [28], studying how the interacting plasmonic resonances in real

dimers will interfere and create non-trivial regimes in their time dynamics. First, we characterize isolated single NPs. Ag-NPs show typically sharp plasmonic resonances and long decay times in their time dynamics for the total dipole moment, whereas Au-NPs show wider resonances and shorter decay times (Figure S3). These single NPs are now combined to build a Ag–Ag Rabi dimer (Figure 2), and a Au–Ag Fano dimer (Figure 3).

Figure 2 shows the Ag–Ag Rabi dimer extinction cross sections under continuous wave (CW) with either a plane wave or with two point dipoles, and the time dynamics under the pulsed excitation. In Figure 2a, the Rabi dimer exhibits the S- and A-modes as plasmonic peaks at 465 and 385 nm, respectively. Those peaks were revealed using 2 fs pulses. In Figure 2b, the time dynamics of the total dipole moment show that d_{tot} oscillates and decays after the initial excitation, showing up to four significant beats for $1 \text{ fs} < t < 40 \text{ fs}$. This means that the plasmon energy becomes transferred back and forth between the NPs, for much longer times than the duration of the initial pulse, eventually decaying. We see that this case is analogous to the Rabi dimer made of the Drude metal (Figure 1c) but, unlike the multiple beats with almost no decay in the Drude

Figure 2: (a) Extinction spectra and surface charge density maps for the Ag–Ag Rabi dimer under CW excitation. We use here two approaches: the electromagnetic wave excitation and the excitation with two anti-symmetric dipoles. In this way, we can see both symmetric and anti-symmetric modes in our Rabi dimer. (b) Dimer's time dynamics under the dipole excitation for a pulse at $\lambda_0 = 465 \text{ nm}$. We used $R_{NP1} = R_{NP2} = 20 \text{ nm}$, gap = 4 nm, and a pulse of $\Delta t = 2 \text{ fs}$ (shown in blue).

Figure 3: Dynamic properties of the Au–Ag Fano dimer. (a) Extinction of the dimer for the electromagnetic CW excitation. Here, we also show the structure of the induced dipoles in the dimer, which leads to the FE. (b) Dimer’s time dynamics under the dipole excitation for a pulse with $\lambda_0 = 589$ nm. We used $R_{Au} = 50$ nm, $R_{Ag} = 5$ nm, $L_{Ag} = 27$ nm, gap = 4 nm, and a pulse of $\Delta t = 3$ fs. The spectrum for the dipole excitation pulse is shown in blue.

case, the Rabi dimer made of a real metal results in a much faster decay, although still showing several beats. This multi-beat behavior is certainly the key feature of the time dynamics of the Rabi-resonance systems. Again, the frequency of the beats in the dynamics in Figure 2b is given by the difference between the frequencies of the modes, $\omega_{A\text{-mode}} - \omega_{S\text{-mode}}$, since these modes (A- and S-modes) dominate the response of the dimer in the frequency domain (Figure 2a).

In the next step, we look at the results for the Au–Ag Fano dimer (Figure 3). Unlike the Rabi bi-harmonic mode, this Fano dimer is multi-harmonic, which is evidenced as a broad spectrum with the destructive interference dip in the extinction spectra at 591 nm (Fano anti-resonance), surrounded by two maxima, at 554 and 618 nm. Dipoles in each NP are schematically shown as black arrows for each resonance. Then, the dimer is excited with a pulsed point dipole, as schematically depicted in Figure 3b, using a 3-fs pulse centered at the Fano anti-resonance (Figure 3a). The oscillation of the total dipole, d_{tot} , shows a single pronounced beat after the pulse, a fundamental difference with respect to the Rabi dimer, which typically shows

several. We note that we do not consider as a beat the first maximum of the envelope function in Figure 3b, since it reflects the excitation pulse. At long times, we see again the decay with the long characteristic tail falling as $\exp[-t/\tau_s]$, like for the Drude case in Figure 1f. In this case with the Au–Ag dimer, we have for the slow decay time: $\tau_1 \sim 10$ fs; this number reflects the plasmonic decay inside the metals. The dynamical behavior in Figure 3b is the signature of the Fano coupling in the time domain. A coupling of this kind exhibits only one beat and leads to the long-time coherent energy dissipation within the Fano dimer.

To summarize our observations regarding the FE in the time and frequency domains, we look now at several related parameters. The most interesting observation is that Fano dynamics have at most only one beat, whereas Rabi dynamics may have virtually any number of beats. In Figure 4a (left), we show the data for an interval limited to 80 fs. The Rabi dynamics for the Drude and Ag NPs have four and seven beats in that interval, respectively (see Figures 1c and 2b). The Au–Ag Fano dimer exhibits only two temporal maxima in the amplitude, from which only one is a beat (Figure 3b). In the Fano dynamics of the

Au–Ag dimer, the first temporal maximum comes simply from the excitation pulse and the second one appears as a beat, due to the coherent energy transfer between NPs (Figure 3b). For the Fano dimers composed of Drude NPs, we do not see beats, however; simultaneously, the dynamic trace contains a very characteristic tail, which is typical for the Fano dynamics (Figure 1f and Figure S12). This fundamental difference between the Rabi and Fano cases comes from the fact that the response of a Rabi system in the frequency domain is bi-harmonic (Figure 1b), whereas the spectrum of a Fano system in the frequency domain is multi-harmonic (Figure 1f). Physically, Rabi dynamics showcased here should be more coherent since it arises in two oscillators with weak decoherence. However, the Fano system should be less coherent since it has one oscillator with very fast decay. Indeed, we observe such property. Remarkably, despite the strong decoherence in the Fano systems, we actually observe their characteristic and prominent coherent dynamics, in the form of the single beat and the slowly decaying tail at long times. The fundamental differences between the dynamical responses of the considered systems can be also seen from the other panels in Figure 4. Here, we need to look at the following parameters: $R_\tau = \tau_{\text{rel, NP2}}/\tau_{\text{rel, NP1}}$, $R_{\text{OS}} = \text{OS}_{\text{NP2}}/\text{OS}_{\text{NP1}}$, and $R_\gamma = \gamma_{\text{NP2}}/\gamma_{\text{NP1}}$, where $\tau_{\text{rel}, i}$, OS_i , and γ_i are the relaxation time, the oscillator strength, and the plasmon resonance broadening, respectively, for the i th NP. In the Rabi model, the above ratios are all around unity, and this property leads to the typical bi-harmonic behavior. In contrast to the Rabi model, the Fano dynamics exhibit fundamentally different spectral properties: $R_\tau \ll 1$, $R_{\text{OS}} \gg 1$, and $R_\gamma \gg 1$ (Figure 4). The above inequalities are the characteristic properties of a Fano system, which should contain one oscillator with a broad resonance and one with a narrow absorption line. The last parameter to consider in Figure 4b is the ratio of the peak extinction cross sections, $R_{\text{peak}} = \sigma_{\text{peak, NP2}}/\sigma_{\text{peak, NP1}}$. Interestingly, R_{peak} can be both small and large in the Fano case. Therefore, the latter parameter cannot be used to discriminate between the Fano and Rabi regimes.

Another property that we observed in our calculations is non-locality of the Fano dynamics. Different responses from the same Fano dimer may show qualitatively different time traces. For example, the coherent beat for the Au–Ag Fano dimer does not appear if we excite the system with a point dipole placed next to the Ag NP (Figure S9). This property also concerns local electric fields in an NP complex. In Figure S10, for the Fano dimer made of Drude metals, we see that the dynamic electric field displays the characteristic coherent beat only inside the NP₂, whereas the fields between the NPs and inside the smaller NP do not show such a feature.

In our study, the Drude NPs serve as convenient physical models, whereas Au and Ag NPs in a liquid matrix represent realistic experimental systems. Our choice of shapes, sizes, and materials in the Fano and Rabi dimers is based on the following considerations: (1) The Rabi dimer is designed with the Ag NPs since silver NPs exhibit a narrow and strong plasmonic resonance. In this case, the resonant interaction of two Ag NPs can be strong, and the resulting collective spectrum can show the Rabi-splitting effect. Indeed, we see this splitting effect in the computed spectra in Figure 2. Furthermore, to obtain a prominent Rabi splitting, the distance between the NPs should be relatively small. We took 4 nm, which is a typical NP–NP distance in the DNA–origami assemblies [29]. (2) The idea behind the design of the Au–Ag Fano dimer in Figure 3 is to use two different materials. The Au NP should be large and should have a broad plasmonic peak (see Figure S3b). Simultaneously, the other element has to have a very narrow plasmonic peak, and the spectral position of this peak should be tunable. A plasmonic NP that satisfies these requirements is a Ag nanorod (see Figure S3c). Its plasmonic resonance can be tuned to match the center of the broad plasmonic peak of the Au NP. This Ag nanorod should be normal to the surface of the Au sphere because this configuration leads to the strongest NP–NP interaction in this hybrid dimer. Again, the NP–NP distance should be taken small enough to have a strong coupling. Using the above ideas and principles, we can design a dimer with the spectrum exhibiting a strong and prominent FE (see Figure 3b). To finish this consideration, one should mention about the possibility to tune the NP–NP gap. The Supporting Information shows such data. In Figure S11, we vary the NP–NP gap in the case of the Drude dimer made of two Drude metals. By increasing the gap, one can see a smooth transition from the Rabi regime to the Fano one. As the system undergoes the Rabi-to-Fano transition, the number of beats in the temporal dynamics drops from two to zero, with the typical slowly decaying tail at long times previously described (see Figure S12).

Theoretically, only a few studies have been published so far on the coherent time dynamics of surface plasmons in NP assemblies [29–32]. Technologically, Fano and Rabi dimers can be fabricated with lithographic techniques [33–35] or using bio-assembly [29]. One powerful tool to observe single NP responses in the frequency domain is dark-field spectroscopy, i.e., the measurement of scattering in the far field. This method was successfully applied to a variety of plasmonic NPs [13, 36–39]. Moreover, this method can be also used in time-resolved studies [39]. In our study, we predict that the Fano dimer should show a unique relaxation pattern in the dynamics of the induced dipole

Figure 4: Summary with several kinetic parameters, for the Fano (red) and Rabi (blue) regimes, for all the dimers presented in this study (schematically shown in the upper row). (a) Temporal properties of the dimer's numbers of beats (left panel), and of the decay-time ratios of isolated NPs (right panel). (b) Ratios of properties of single isolated NPs in the frequency domain, for the oscillator strength (top-left panel), the plasmon extinction broadenings (top-right panel) and the extinction maxima (bottom panel).

(Figures 1f and 3b, Figures S8 and S9), a behavior that can be observed with time-resolved far-field scattering. The point-dipole excitation, used in our study to demonstrate the time dynamics and the related NP–NP energy transfer, can be realized experimentally with plasmonic tip-based spectroscopies, such as Nanoscopy [40] or Scanning Near-field Optical Microscopy (SNOM) [41]. The time resolution, which is needed to observe the predicted Fano dynamics, is in the range of 5–10 fs. Simultaneously, the majority of the current experiments in the field of ultrafast spectroscopy have been performed with the time resolution of ~80 fs [13, 42–44], which is not sufficient for our effects. However, the area of time-resolved spectroscopies develops fast and experiments with a very high resolution, in the range of ~1–15 fs already exist [39, 45–48]. Along with the all-optical

approaches cited above, one should mention a proposal on sub-fs electron-beam microscopy [32] that can also be applied to observe the coherent plasmonic dynamics described in our study.

When considering future applications, the field of nanostructures is unique since it allows us to realize in practice physical models of our choice. As mentioned above, potential experiments on the Fano dynamics of plasmonic excitations can be performed either with lithographic samples or with DNA-assembled NP complexes. The latter case offers more precise control over the inter-particle gaps [29]. However, the lithographic systems can be more uniform and, therefore, would not require single-particle measurements. In our plasmonic dimers, the Fano and Rabi effects come from the interference of localized

excitations. Furthermore, the Fano-like effects and their time-dependent manifestations can also appear in purely photonic systems, which couple strongly with far fields [49]. Another important point we need to make is that our predictions relate to the plasmonic systems described by classical electrodynamics. In other words, our models are classical. The Fano and Rabi effects in semiconductor quantum dots and other quantum systems require very different treatments, which are based on quantum mechanics. Therefore, such systems should be studied separately, and our classical results may not apply to them.

4 Conclusions

We have studied the time-domain dynamics of plasmonic dimers, with the emphasis on the Fano and Rabi regimes. Whereas single NPs show trivial time dynamics and fast decays with no coherent energy transfer (i.e., no beats), both kinds of dimers show non-trivial decay dynamics with notable features due to coherent energy transfer. The fundamental difference between the time dynamics of the Rabi and Fano systems is the number of beats. The Rabi system may have any number of beats, whereas the Fano system may have at most one. Another very characteristic feature of the Fano dimer is a prominent long-lasting tail in its response. Moreover, the peculiar dynamical properties of the strongly interacting dimers are related to the spectral properties of the isolated NPs constituting the dimers. Our results could be tested with already available structures and the rapidly developing field of ultra-fast spectroscopies. As such, we expect that our results will motivate further research within temporal coherent plasmonics.

Acknowledgments: O.A.-O. and A.O.G. were supported by the Nanoscale & Quantum Phenomena Institute at Ohio University. L.V.B. was supported by the Institute of Fundamental and Frontier Sciences, University of Electronic Science and Technology of China and China Postdoctoral Science Foundation (2017M622992 and 2019T120820). Z.M.W. was funded by the National Key Research and Development Program (2019YFB2203400), the “111 Project” (B20030), and the UESTC Shared Research Facilities of Electromagnetic Wave and Matter Interaction (Y0301901290100201).

Author contribution: All the authors have accepted responsibility for the entire content of this submitted manuscript and approved submission.

Research funding: None declared.

Conflict of interest statement: The authors declare no conflicts of interest regarding this article.

References

- [1] U. Fano, “Effects of configuration interaction on intensities and phase shifts,” *Phys. Rev.*, vol. 124, pp. 1866–1878, 1961.
- [2] A. E. Miroshnichenko, S. Flach, and Y. S. Kivshar, “Fano resonances in nanoscale structures,” *Rev. Mod. Phys.*, vol. 82, pp. 2257–2298, 2010.
- [3] J. Chen, F. Gan, Y. Wang, and G. Li, “Plasmonic sensing and modulation based on Fano resonances,” *Adv. Optical Mater.*, vol. 6, p. 1701152, 2018.
- [4] G. P. Wiederrecht, G. A. Wurtz, and J. Hranisavljevic, “Coherent coupling of molecular excitons to electronic polarizations of noble metal nanoparticles,” *Nano Lett.*, vol. 4, pp. 2121–2125, 2004.
- [5] W. Zhang, A. O. Govorov, and G. W. Bryant, “Semiconductor-metal nanoparticle molecules: hybrid excitons and the nonlinear Fano effect,” *Phys. Rev. Lett.*, vol. 97, p. 146804, 2006.
- [6] R. A. Shah, N. F. Scherer, M. Pelton, and S. K. Gray, “Ultrafast reversal of a Fano resonance in a plasmon-exciton system,” *Phys. Rev. B*, vol. 88, p. 075411, 2013.
- [7] J. B. Lassiter, H. Sobhani, M. W. Knight, W. S. Mielczarek, P. Nordlander, and N. J. Halas, “Designing and deconstructing the Fano lineshape in plasmonic nanoclusters,” *Nano Lett.*, vol. 12, pp. 1058–1062, 2012.
- [8] G. Bachelier, I. Russier-Antoine, E. Benichou, et al., “Fano profiles induced by near-field coupling in heterogeneous dimers of gold and silver nanoparticles,” *Phys. Rev. Lett.*, vol. 101, p. 197401, 2008.
- [9] Z.-J. Yang, Z.-S. Zhang, W. Zhang, Z.-H. Hao, and Q.-Q. Wang, “Twinned Fano interferences induced by hybridized plasmons in Au–Ag nanorod heterodimers,” *Appl. Phys. Lett.*, vol. 96, p. 131113, 2010.
- [10] N. W. Bigelow, A. Vashillo, J. P. Camden, and D. J. Masiello, “Signatures of Fano interferences in the electron energy loss spectroscopy and cathodoluminescence of symmetry-broken nanorod dimers,” *ACS Nano*, vol. 7, pp. 4511–4519, 2013.
- [11] A. Nazir, S. Panaro, R. Proietti Zaccaria, C. Liberale, F. De Angelis, and A. Toma, “Fano coil-type resonance for magnetic hot-spot generation,” *Nano Lett.*, vol. 14, no. 6, pp. 3166–3171, 2014.
- [12] L. Khosravi Khorashad, L. V. Besteiro, Z. Wang, J. Valentine, and A. O. Govorov, “Localization of excess temperature using plasmonic hot spots in metal nanostructures: combining nano-optical antennas with the Fano effect,” *J. Phys. Chem. C*, vol. 120, pp. 13215–13226, 2016.
- [13] A. Lombardi, M. P. Grzelczak, E. Pertreux, et al., “Fano interference in the optical absorption of an individual Gold–Silver nanodimer,” *Nano Lett.*, vol. 16, pp. 6311–6316, 2016.
- [14] S. Simoncelli, Y. Li, E. Cortés, and S. A. Maier, “Imaging plasmon hybridization of Fano resonances via hot-electron-mediated absorption mapping,” *Nano Lett.*, vol. 18, pp. 3400–3406, 2018.
- [15] J. Zhang and A. Zayats, “Multiple Fano resonances in single-layer nonconcentric core–shell nanostructures,” *Opt. Express*, vol. 21, pp. 8426–8436, 2013.
- [16] M. Kroner, A. O. Govorov, S. Remi, et al., “The nonlinear Fano effect,” *Nature*, vol. 451, pp. 311–314, 2008.
- [17] M. L. Kerfoot, A. O. Govorov, D. Lu, et al., “ptophonics with coupled quantum dots,” *Nat. Commun.*, vol. 5, p. 3299, 2014.

- [18] Y. I. Mazur, V. G. Dorogan, D. Guzun, et al., “Measurement of coherent tunneling between InGaAs quantum wells and InAs quantum dots using photoluminescence spectroscopy,” *Phys. Rev. B*, vol. 82, p. 155413, 2010.
- [19] J. Bellessa, C. Bonnand, J. C. Plenat, and J. Mugnier, “Strong coupling between surface plasmons and excitons in an organic semiconductor,” *Phys. Rev. Lett.*, vol. 93, p. 036404, 2004.
- [20] J. Dintinger, S. Klein, F. Bustos, W. L. Barnes, and T. W. Ebbesen, “Strong coupling between surface plasmon-polaritons and organic molecules in subwavelength hole arrays,” *Phys. Rev. B*, vol. 71, p. 035424, 2005.
- [21] T. Yoshie, A. Scherer, J. Hendrickson, et al., “Vacuum Rabi splitting with a single quantum dot in a photonic crystal nanocavity,” *Nature*, vol. 432, pp. 200–203, 2004.
- [22] H. Yokoyama and K. Ujihara, *Spontaneous Emission and Laser Oscillation in Micro-cavities*, 1st ed. Cleveland, OH, USA, CRC Press, 1995.
- [23] P. Nordlander, C. Oubre, E. Prodan, K. Li, and M. I. Stockman, “Plasmon hybridization in nanoparticle dimers,” *Nano Lett.*, vol. 4, pp. 899–903, 2004.
- [24] J. R. Schaibley, A. P. Burgers, G. A. McCracken, et al., “Direct detection of time-resolved Rabi oscillations in a single quantum dot via resonance fluorescence,” *Phys. Rev. B*, vol. 87, p. 115311, 2013.
- [25] S. K. Rajendran, W. Wang, D. Brida, et al., “Direct evidence of Rabi oscillations and antiresonance in a strongly coupled organic microcavity,” *Phys. Rev. B*, vol. 91, p. 201305, 2015.
- [26] K. Kuruma, Y. Ota, M. Kakuda, S. Iwamoto, and Y. Arakawa, “Time-resolved vacuum Rabi oscillations in a quantum-dot–nanocavity system,” *Phys. Rev. B*, vol. 97, p. 235448, 2018.
- [27] S. A. Maier, *Plasmonics: Fundamentals and Applications*, 1st ed. New York, NY, USA, Springer, 2007.
- [28] P. B. Johnson and R. W. Christy, “Optical constants of the noble metals,” *Phys. Rev. B*, vol. 6, pp. 4370–4379, 1972.
- [29] E.-M. Roller, L. V. Besteiro, C. Pupp, L. K. Khorashad, A. O. Govorov, and T. Liedl, “Hotspot-mediated non-dissipative and ultrafast plasmon passage,” *Nat. Phys.*, vol. 13, pp. 761–765, 2017.
- [30] C. M. Lazzarini, L. Tadzio, J. M. Fitzgerald, J. A. Sánchez-Gil, and V. Giannini, “Linear ultrafast dynamics of plasmon and magnetic resonances in nanoparticles,” *Phys. Rev. B*, vol. 96, p. 235407, 2017.
- [31] R. Faggiani, A. Losquin, J. Yang, E. Mårssell, A. Mikkelsen, and P. Lalanne, “Modal analysis of the ultrafast dynamics of optical nanoresonators,” *ACS Photonics*, vol. 4, pp. 897–904, 2017.
- [32] M. I. Stockman, M. F. Kling, U. Kleineberg, and F. Krausz, “Attosecond nanoplasmonic-field microscope,” *Nat. Photonics*, vol. 1, pp. 539–544, 2007.
- [33] S. A. Maier, P. G. Kik, H. A. Atwater, et al., “Local detection of electromagnetic energy transport below the diffraction limit in metal nanoparticle plasmon waveguides,” *Nat. Mater.*, vol. 2, pp. 229–232, 2003.
- [34] N. Mahi, G. Lévêque, O. Saison, et al., “In depth investigation of lattice plasmon modes in substrate-supported gratings of metal monomers and dimers,” *J. Phys. Chem. C*, vol. 121, pp. 2388–2401, 2017.
- [35] Y. Zhang, G. Demesy, M. Haggui, et al., “Nanoscale switching of near-infrared hot spots in plasmonic oligomers probed by two-photon absorption in photopolymers,” *ACS Photonics*, vol. 5, pp. 918–928, 2018.
- [36] J. H. Hodak, A. Henglein, and G. V. Hartland, “Photophysics of nanometer sized metal particles: electron-phonon coupling and coherent excitation of breathing vibrational modes,” *J. Phys. Chem. B*, vol. 104, pp. 9954–9965, 2000.
- [37] C. Sönnichsen, T. Franzl, T. Wilk, et al., “Drastic reduction of plasmon damping in gold nanorods,” *Phys. Rev. Lett.*, vol. 88, p. 077402, 2002.
- [38] H. Baida, D. Mongin, D. Christofilos, et al., “Ultrafast nonlinear optical response of a single gold nanorod near its surface plasmon resonance,” *Phys. Rev. Lett.*, vol. 107, p. 057402, 2011.
- [39] M. Zavelani-Rossi, D. Polli, S. Kochtcheev, et al., “Transient optical response of a single gold nanoantenna: the role of plasmon detuning,” *ACS Photonics*, vol. 2, pp. 521–529, 2015.
- [40] A. Giugni, B. Torre, M. Allione, et al., “Experimental route to scanning probe hot-electron nanoscopy (HENs) applied to 2D material,” *Adv. Opt. Mater.*, vol. 5, p. 1700195, 2017.
- [41] H. Heinzelmann and D. W. Pohl, “Scanning near-field optical microscopy,” *Appl. Phys. A*, vol. 59, pp. 89–101, 1994.
- [42] S. Link and M. A. El-Sayed, “Spectral properties and relaxation dynamics of surface plasmon electronic oscillations in gold and silver nanodots and nanorods,” *J. Phys. Chem. B*, vol. 103, pp. 8410–8426, 1999.
- [43] G. V. Hartland, “Optical studies of dynamics in noble metal nanostructures,” *Chem. Rev.*, vol. 111, pp. 3858–3887, 2011.
- [44] H. Harutyunyan, A. B. F. Martinson, D. Rosenmann, et al., “Anomalous ultrafast dynamics of hot plasmonic electrons in nanostructures with hot spots,” *Nat. Nanotechnol.*, vol. 10, pp. 770–774, 2015.
- [45] R. Mittal, R. Glenn, I. Saytashev, V. V. Lozovoy, and M. Dantus, “Femtosecond nanoplasmonic dephasing of individual silver nanoparticles and small clusters,” *J. Phys. Chem. Lett.*, vol. 6, pp. 1638–1644, 2015.
- [46] G. Della Valle, M. Conforti, G. Cerullo, and D. Brida, “Real-time optical mapping of the dynamics of nonthermal electrons in thin gold films,” *Phys. Rev. B*, vol. 86, p. 155139, 2012.
- [47] A. Kubo, Y. Suk Jung, H. Koo Kim, and H. Petek, “Femtosecond microscopy of localized and propagating surface plasmons in silver gratings,” *J. Phys. B At. Mol. Opt. Phys.*, vol. 40, p. S259, 2007.
- [48] P. Vasa, W. Wang, R. Pomraenke, et al., “Real-time observation of ultrafast Rabi oscillations between excitons and plasmons in metal nanostructures with J-aggregates,” *Nat. Photonics*, vol. 7, pp. 128–132, 2013.
- [49] S. E. Svyakhovskiy, V. V. Ternovski, and M. I. Tribelsky, “Anapole: its birth, life, and death,” *Optic Express*, vol. 27, p. 23894, 2019.

Supplementary Material: The online version of this article offers supplementary material (<https://doi.org/10.1515/nanoph-2020-0229>).