Nonlinear optical response of a local surface plasmon coupled to a 2D material

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We present a theoretical study of the optical response of a nonlinear oscillator formed by coupling a metal nanoparticle local surface plasmon resonance to excitonic degrees of freedom in a monolayer transition-metal dichalcogenide. We show that the combined system should exhibit strong anharmonicity in its low-lying states, predicting for example a seven order-of-magnitude increase in nonlinearity relative to a silicon photonic crystal cavity. Arrays of such nanoscale nonlinear oscillators could be used to realize novel optical metamaterials; alternatively, an individual nanoparticle-monolayer construct could be coupled to an optical resonator to mediate efficient input-output coupling to propagating fields.

A localized surface plasmon resonance (LSPR) of a metal nanoparticle (MNP) may be modeled as a quantized harmonic oscillator [1]. For a given resonance frequency, the energy states form a harmonic ladder broadened by Ohmic loss of the metal [2]. The LSPR field is confined within a volume slightly larger than the MNP, which is significantly smaller than the free-space wavelength of light at the LSPR resonant frequency [3]. In this Letter, we analyze a scheme for turning an LSPR into a quantum nonlinear resonator via near-field coupling to an atomically thin 2D material, such as a transition metal dichalcogenide (TMD). Two key considerations motivate this proposal: that monolayer TMDs can provide strong Kerr nonlinearity with a favorable coherence-to-decoherence ratio [4], and that monolayer TMDs are sufficiently thin to overlap substantially with the nanoscale LSPR evanescent field.

There are many complementary schemes for inducing nonlinearity in an optical oscillator [5–7]. In the (arguably) most widely applicable approach, a Kerr-nonlinear medium can be introduced into an otherwise passive optical cavity. If the ratio of nonlinear Kerr effect to absorption loss is sufficiently large, such a system may be useful for quantum optics and quantum information processing. Unfortunately, it is difficult to find materials with a large nonlinearity-to-loss ratio, which are amenable to incorporation within conventional or nanofabricated optical resonators. For example, although some bulk materials such as silicon have a significant Kerr optical nonlinearity at near-infrared optical frequencies, they also have a large imaginary nonlinear response (two-photon absorption) that gives rise to comparably large losses [8]. Setting aside the issue of losses, nanophotonic resonators incorporating conventional optical materials such as silicon generally do not possess a sufficiently large Kerr nonlinearity at low photon numbers for quantum information processing applications.

Monolayer MoS$_2$ has an order-of-magnitude larger real $\chi^{(3)}$ than bulk silicon and can have a significant real-to-imaginary ratio of $\chi^{(3)}$ [4]. Its 2D structure furthermore makes it a promising candidate to induce a large coherent nonlinearity through direct coupling to the strongly confined field of an LSPR. Such an LSPR-TMD system could be efficiently coupled to optical input-output modes by incorporation within an optical cavity, in a manner analogous to the way that a Josephson-Junction nonlinear LC oscillator can be coupled to propagating fields by a microwave resonator. With regard to the potential scalability of such a device concept, we note that, while automated assembly of the type of system we envision could be challenging to realize, the material components we require may be producible in bulk by chemical MNP synthesis [9] and chemical-vapor-deposition (CVD) growth of TMDs [10, 11]. One can alternatively envision a large number of MNPs dispersed on a TMD membrane to form an array of nonlinear oscillators with complex couplings in both real and frequency spaces; such an optical metamaterial could provide rich nonlinear dynamics for reser-
vior computing-type architectures [12].

Let us consider an MNP driven by a classical light source. A monolayer TMD is positioned nearby the MNP so that the LSPR field has a considerably large overlap with the monolayer TMD (see Fig. [1]). To understand the system, we adopt the quantum master equation [13, 14], which is particularly suitable for optical frequencies. In this framework, the unitary evolution of the system is modeled through a Hamiltonian $H = H_a + H_{\text{drive}}$ where $H_a$ and $H_{\text{drive}}$ are linear harmonic ladders with equal level spacing of $\omega_a/2$ (see the supplemental material [19] for details):

$$H_a = \hbar\omega_a a^\dagger a + \hbar\chi a^\dagger a a,$$

$$H_{\text{drive}} = -i\hbar\sqrt{\kappa}(a^\dagger r e^{-i\omega t} - a r^* e^{i\omega t}),$$

where $\omega_a$ is the resonant frequency of the LSPR field, $\chi$ is the Kerr nonlinear coefficient, and $a, a^\dagger$ are the annihilation and creation operators of the LSPR field. Here, $\kappa$ is the coupling coefficient, $\epsilon$ is the drive, and $\omega$ is the frequency of the drive field. The nonunitary evolution of the system undergoes dissipation through the Lindblad operators $L_1 = \sqrt{\gamma}a$ with $\gamma = \gamma_r + \gamma_o$, which includes both the radiative ($\gamma_r$) and nonradiative ($\gamma_o$) decays where the nonradiative Ohmic loss is dominant ($\gamma \simeq \gamma_o \gg \gamma_r$) [2], and $L_2 = \sqrt{\chi/2}a a$, which is the nonlinear loss (two-photon absorption) through the coupling with the nonlinear 2D material. As a consequence, the effective energy levels appear as in Fig. [1]. Without nonlinear coupling to the 2D material, the energy levels for a given frequency $\omega_a$ are a linear harmonic ladder with an equal level spacing of $\hbar\omega_a$, with a level broadening $\hbar\gamma$. The nonlinearity modifies the energy levels in two ways: both the energy level shift and the level broadening vary with LSPR photon number, resulting in an anharmonic energy ladder. Assuming a single-frequency optical driving field, it is apparent that, in order not to excite an effectively linear (trivial) response, the nonlinear energy shifts must be larger than the level broadenings.

The nonlinear energy shift and broadening depend on the coefficients $\chi$ and $\chi'$ (both real-valued), which are calculated using the results in [15, 16, 19, 21] (see the detailed derivation in the supplemental material [19]):

$$\chi + i\chi' = \frac{27\epsilon_0\epsilon_a^2}{4} \int d^3 r \chi^{(3)}(r)|f_0(r)|^4,$$

where $\epsilon_0$ is the vacuum permittivity and $\chi^{(3)}(r)$ is the position ($r$) dependent third-order nonlinear susceptibility. Since monolayer TMDs have a negligibly small optical response to out-of-plane-polarized electric fields [22, 23], we count only the in-plane LSPR field component (with respect to the monolayer 2D material): $f_0(r)$ is the in-plane component of the LSPR mode function $f(r)$, normalized as $\int d^3 r \epsilon_0\epsilon(r)|f_0(r)|^2 = 1$ with the position dependent relative permittivity $\epsilon_r(r)$. Evidently, the overlap between the LSPR mode field and the 2D material volume must be large to increase $\chi$. We note that a large overlap also increases the nonlinear broadening of two-photon absorption. Hence, the the ratio $\chi/\chi'$ must be large, which depends in general on the material and the operating frequency.

To appreciate how difficult it is to make a practical nonlinear (anharmonic) oscillator, let us consider an optical resonator filled with silicon. We make an example of the state-of-the-art smallest photonic crystal cavity to increase the nonlinear energy shift: a cavity of volume $V_c = 0.02 \mu m^3$ having a cavity $Q = 2 \times 10^5$ [24]. Then, using $\text{Re}\chi^{(3)} \sim 4.8 \times 10^{-20} m^2/V^2$ of silicon at 1 eV photon frequency (energy) [8], Eq. (3) leads to $\hbar\chi \sim 1 \times 10^{-9} eV$. However, the linear cavity-lifetime-induced level broadening (ignoring the intrinsic linear loss of silicon) is $\hbar\gamma \sim 5 \times 10^{-6} eV$. Hence, the line broadening is dominant over the nonlinear energy level shift at low photon number. Although quantum well materials such as GaAs/InGaAs have appreciable Kerr nonlinearity ($\text{Re}\chi^{(3)} = 1.3 \times 10^{-19} m^2/V^2$ at 1.3 $\mu m$ wavelength [25]), the two-photon absorption is large ($\text{Im}\chi^{(3)} = 1.4 \times 10^{-18} m^2/V^2$ [24]) and the linear absorption is excessively large (for $\alpha > 10 \ cm^{-1}$) [26, 27] so that the nonlinearity-to-broadening ratio is even less favourable than our silicon example. Chalcogenide glass materials are not much better due to the same reason of large linear and two-photon absorption [28].

We will show below that our proposed system could achieve few-photon nonlinear energy shifts that dominate broadening, and thus realize a quantum nonlinear oscillator. We first discuss several design aspects for the system. We utilize the large ratio of the real and the imaginary values of $\chi^{(3)}$ in a monolayer MoS$_2$ by detuning slightly from the two-photon resonant frequency 1.06 eV [4]. We must also design an MNP to support an LSPR resonance near 1.06 eV with a sufficiently small $\hbar\gamma$. The resonance frequency $\omega_a$ of the LSPR mode is well known to depend upon the geometry of the MNP [29, 31], with thin MNPs exhibiting lower resonance frequencies [32]. The reason is that the portion of the LSPR field inside the metal reduces as the metal layer becomes thinner, which tends to red-shift the resonance frequencies [31].

We need to maximize the in-plane component of the LSPR field that overlaps the monolayer MoS$_2$. A natural choice of geometry is a disk shape MNP with a large ratio of diameter to thickness. A large diameter improves the in-plane LSPR field component while a thinner disk shape pull $\omega_a$ down from the nominally visible photonic resonances of silver. Using finite-element-method (FEM) software (COMSOL), we found that a diameter-to-thickness ratio of 22 for a silver disk should provide an LSPR resonance frequency near the target. On the other hand, the value of $\gamma$ of an LSPR is known to be independent of shape and size, since it is completely determined by the complex dielectric function of the material; once the material and operating frequency are determined, there is not much one can do to adjust $\gamma$ [31]. For this theoretical study, we take from the work of Wang and Shen a Q-factor $Q = \omega_a/\gamma = 50$ [31]. In reality, the
surface imperfections may reduce the Q-factor.

We also need to consider potential impacts on the intrinsic optical properties of a TMD monolayer when an MNP is placed nearby—an excessively small gap between the two may alter the band structure of the 2D material and impact the nonlinearity adversely. To address this problem, we consider a tight binding band Hamiltonian

\[ H_{\text{band}} = \sum_{\langle i,j \rangle} J_{ij} c_i^\dagger c_j + \sum_i L_i c_i^\dagger c_i + \sum_i M_i (c_i^\dagger d_i + c_i d_i^\dagger) \]

where \( c_i^\dagger, d_i \) are the second-quantized fermionic creation operators for the electrons in the 2D material lattice and the impurity nearby, respectively. The first term is the nearest-neighbor (nn) hopping energy, the second is the on-site energy, and the third is the interaction energy between the 2D material and the impurity sites. We note that \( J_{ij} \) and \( M_i \) originate from the Coulomb potential. It is customary to ignore the next-nearest neighborhood (nnn) hopping since the matrix element for this remote potential is negligibly small compared to the nearest-neighbor hopping. Hence, to preserve the band structure intact, we require the condition \( |M_i| \ll |J_{ij}| \), which can be fulfilled if the distance between the 2D lattice sites and the impurity sites becomes larger than the next-nearest neighbor (nnn) distance of the 2D lattice. When the hexagonal atomic arrangement of monolayer TMDs is considered, the nnn distance is 0.32 nm (MoS\(_2\)), approximately 1.7 times longer than nn. Indeed, Liu et al. recently discussed the encapsulation of monolayer TMDs and found that using a few-layer hexagonal boron-nitride (hBN) buffer preserved the properties of monolayers successfully. Man et al. discussed the successful protection of monolayer properties using even a single layer hBN buffer. Hence, in this theoretical paper, we set the minimum distance between the monolayer TMD and the MNP as 0.3 nm, corresponding to the thickness of a single layer hBN. Additionally, since the operating frequency is detuned from both the one- and two-photon excitonic resonance of the monolayer TMD, exciton generation will be minimal and we neglect metal-dielectric effects such as mirror charges.

To understand the dynamics of the system, we consider the adjoint master equation for a Heisenberg picture operator \( \hat{Q} \) as [14]:

\[ \dot{Q} = -i \frac{\hbar}{\gamma} [Q, H] + \sum_j \left( L_j^\dagger Q L_j - \frac{1}{2} L_j^\dagger L_j Q - \frac{1}{2} Q L_j^\dagger L_j \right) . \]  

Then, we easily obtain the dynamical equation for the LSPR field using the Hamiltonian in Eq. 1 and 2 and the Lindblad operators:

\[ \dot{a} = -i \Delta_a - i \chi (a^\dagger a) a - \frac{\gamma}{2} a - \frac{\chi'}{2} (a^\dagger a) a - \sqrt{\kappa} \xi, \]  

where \( \Delta_a = \omega_a - \omega \). In the limit of considering only the lowest three energy levels with a moderate drive power, the steady-state population in the second excited level can be expressed using the population in the first level as (supplemental material [19]):

\[ \rho_{22,ss} = \frac{4 \kappa \varepsilon^2 / (\gamma + \chi')^2}{1 + (4 \kappa \varepsilon^2 / (\gamma + \chi')^2)^2} \rho_{11,ss}. \]

Note that for a linear system (\( \chi = \chi' = 0 \)), the population is \( \rho_{22,ss} = (4 \kappa \varepsilon^2 / \gamma^2) / (1 + 4 \kappa \varepsilon^2 / \gamma^2) \), which exhibits the conventional saturation feature with respect to the drive power. The nonlinearity (anharmonicity) kicks in if the quantity \( 4 \chi^2 / (\gamma + \chi')^2 \) is sufficiently large to suppress \( \rho_{22,ss} \). Hence, we should adjust the system design parameters to maximize the nonlinear figure of merit (FOM) \( 4 \chi^2 / (\gamma + \chi')^2 \).

The FOM depends mainly on two design parameters: the MNP size and the drive frequency detuning. The MNP size determines the overlap between the monolayer TMD and the LSPR field, scaling \( \chi \) and \( \chi' \) in the same fashion. A large MNP has a more delocalized field outside the MNP and, thus, the relative portion overlapping the thin nonlinear 2D material is small. On the other hand, detuning of the drive frequency adjusts the ratio between \( \chi \) and \( \chi' \). The values \( \text{Im}[\chi^{(3)}] \) of a monolayer MoS\(_2\) roughly follows a typical Lorentzian lineshape while \( \text{Re}[\chi^{(3)}] \) is the frequency-derivative of \( \text{Im}[\chi^{(3)}] \). Hence, FOM is expected to be nearly zero at the two-photon resonance of the monolayer TMD exciton while the maximum occurs an appropriately detuned frequency.

To maximize the overlap, we place two monolayer MoS\(_2\) planes sandwiching the disk-shaped MNP, with hBN buffers separating the MNP from each of the monolayers. Using an FEM software package (COMSOL), we calculate the detailed field distribution of a given MNP design and subsequently determine \( \chi, \chi' \) according to Eq. 8. Fig. 2 shows the calculated FOM as a function of the silver disk MNP’s diameter (while fixing the diameter-to-thickness ratio at 22) and the drive
frequency detuning from the monolayer MoS$_2$’s two-photon resonance at 1.06 eV. We note that with the fixed diameter-to-thickness ratio of 22, the MNP diameter must be larger than 4 nm so that the MNP disk thickness is larger than the atomic size of silver. In fact, 0.2 nm thin silver shell was experimentally used in earlier work [37]. Remarkably the nonlinear FOM is larger than unity over a wide range of parameters; the unity FOM value is the on-set for a system to be nonlinear. The maximum FOM of 10.7 occurs at a detuning of $-11$ meV with the smallest MNP diameter of 4 nm. The real and the imaginary values of $\chi^{(3)}$ of the monolayer MoS$_2$ at this detuning are $4.0 \times 10^{-19}$ m$^2$/V$^2$ and $1.0 \times 10^{-19}$ m$^2$/V$^2$, respectively [38]. The subsequent $h\chi$ and $h\chi'$ values are obtained as 53 meV and 13 meV, respectively. It is noteworthy that the improvement of FOM by placing additional stacks of monolayer MoS$_2$ with gap-buffers is negligible due to the fast decaying LSPR mode field.

Nonlinear behavior of the system can be clearly seen through the population distribution among energy eigenstates when driven by an external field. For this, we solved the dynamic equation (5) by setting $\sqrt{\kappa\varepsilon} = 10$ meV driving on resonance ($\omega = \omega_s$). The steady-state population distributions are shown in Fig. 3. The linear system ($h\chi = h\chi' = 0$ meV) shows a typical coherent state population distribution, while the nonlinear system ($h\chi, h\chi', h\gamma = 53, 13, 21$ meV) has suppressed population in the upper levels. The population suppression agrees with the above analysis (Eq. (6)). We adjusted the drive power slightly to normalize $\rho_{11}$ since the same drive power will lead to slightly different $\rho_{11}$ populations between the linear and the nonlinear systems. The residual population $\rho_{22}$ in the nonlinear system is caused by the saturation of $\rho_{11}$, which could be effectively further reduced using a smaller drive power.

Another way to see the nonlinearity of the system is to calculate the two-photon correlation $g^{(2)}(\tau) = \lim_{t \to +\infty} \langle a(t)a(t + \tau) a(t + \tau) a(t) \rangle / \langle a(t)a(t) \rangle^2$ [39]. A linear system such as an empty optical cavity will produce $g^{(2)}(\tau) = 1$ for all $\tau$ when driven by a classical source, which corresponds to a coherent state. However, a strongly coupled atom–cavity system exhibits photon blockade and, thus, $g^{(2)}(0)$ close to zero [39, 10]. We obtained the two-photon correlation $g^{(2)}(\tau)$ as shown in Fig. 4 by solving the dynamic equation (Eq. (5)). As expected, a linear system ($\chi = \chi' = 0$) shows a flat $g^{(2)}(\tau) = 1$ for all $\tau$, regardless of the drive power. In contrast, our system shows a strong nonlinearity having $g^{(2)}(0) < 1$. Obviously $g^{(2)}(0)$ depends on the population in the upper excited levels and, hence, it depends on the drive power. When weakly driven, the $g^{(2)}(0)$ value reaches as low as 0.15, predicting strong antibunching in optical fields escaping from the LSPR. As the drive power increases, the upper level starts being populated, degrading $g^{(2)}(0)$ as shown.

We have demonstrated a nonlinear oscillator composed of an LSPR with added Kerr nonlinearity via coupling to monolayer MoS$_2$. To qualify as a quantum nonlinear oscillator, the nonlinear FOM $4\chi^2/\chi'^2$ must be larger than unity. The nonlinearity of a state-of-the-art smallest photonic-crystal point defect cavity filled with silicon is inadequate (FOM $\sim 10^{-6}$), whereas our proposed system has predicted FOM $\sim 10$, which is a seven order-of-magnitude improvement. The improvement stems from the strong field enhancement provided by a metal nanoparticle LSPR and direct coupling to a highly nonlinear monolayer TMD material, which is thin enough to overlap well with the tightly confined LSPR evanescent field. (Although graphene possesses an order-of-magnitude larger real $\chi^{(3)}$ at the same optical frequency than monolayer TMDs, the unfavorable real-to-imaginary $\chi^{(3)}$ ratio of graphene spoils the potential advantage [38], making graphene a less desirable material than the monolayer TMDs.) Moreover, by appropriately detuning the drive frequency, one can maximize the nonlinear FOM thanks to the discrete nonlinear susceptibility spectrum of the monolayer TMDs. It is possible that the practically achievable LSPR loss rate $\gamma$ could be larger than the theoretical value of 21 meV that we used in our calculation, for example, due to surface impurities,

![FIG. 3: Comparison of the steady-state excited level populations when driven by an external field at resonance between a linear and a nonlinear system.](image)

![FIG. 4: Two-photon correlation $g^{(2)}(\tau)$ of linear (L) and nonlinear systems (NL) with various drive powers.](image)
and in this case, the FOM would degrade accordingly. Even in the weakly nonlinear regime, however, pulse sequences analogous to those designed for suppression of population outside the qubit subspace in microwave anharmonic oscillators \cite{11} could be used to enable manipulation of the LSPR state in an effectively nonlinear fashion. Our proposed MNP-TMD quantum nonlinear oscillator could be further coupled to an optical cavity, facilitating its use to construct nonlinear quantum photonic devices. Recent studies demonstrated a decoherence-free operation of a system combining an LSPR mode and an emitter \cite{10, 12}.

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