Interaction-induced photon blockade using an atomically thin mirror embedded in a microcavity

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Narrow dark resonances associated with electromagnetically induced transparency play a key role in enhancing photon-photon interactions. The schemes realized to date relied on the existence of long-lived atomic states with strong van der Waals interactions. Here, we show that by placing an atomically thin semiconductor with ultra-fast radiative decay rate inside a 0D cavity, it is possible to obtain narrow dark- or bright resonances in transmission whose width could be much smaller than that of the cavity and bare exciton decay rates. While breaking of translational invariance places a limit on the width of the dark resonance width, it is possible to obtain a narrow bright resonance that is resilient against disorder by tuning the cavity away from the excitonic transition. Resonant excitation of this bright resonance yields strong photon antibunching even in the limit where the interaction strength is arbitrarily smaller than the non-Markovian disorder broadening and the radiative decay rate of the bare exciton. Our findings suggest that atomically thin semiconductors could pave the way for realization of strongly interacting photonic systems in the solid-state.

Monolayers of transition metal dichalcogenides (TMD) such as MoSe$_2$ or WSe$_2$ constitute a new class of two dimensional (2D) direct band-gap semiconductors [1–4]. Lowest energy elementary optical excitations in TMDs in the absence of free electrons or holes are excitons with an ultra-large binding energy of $\sim 0.5$ eV [5]. Remarkably, recent experiments have demonstrated predominantly spontaneous emission limited exciton transition linewidths in clean MoSe$_2$ or WSe$_2$ flakes that are either suspended or embedded in hexagonal boron nitride (hBN) layers. Since radiative broadening can dominate over disorder induced inhomogeneous broadening, clean TMD monolayers can be considered as ideal two-dimensional (2D) optical materials, realizing atomically thin mirrors [6–8].

In this Letter, we show that it is possible to obtain strong photon blockade effect [9–10] by placing a TMD monolayer inside a 2D microcavity, even in the limit where the cavity and the exciton radiative decay rate, as well as the disorder broadening of the exciton is much larger than the exciton-exciton interaction. When the exciton is resonant with a low quality (Q) factor cavity mode, the coupled system exhibits a dark resonance: the minimum achievable width of this dark transmission window is limited only by non-radiative exciton line broadening. In contrast, when the exciton resonance is red detuned from the cavity mode, the linewidth of the transmission peak associated with the lower polariton can be much smaller than the exciton disorder broadening as well as the radiative decay rate of the bare exciton and cavity modes. Hence, in this limit, the exciton-exciton interaction strength required to observe large photon antibunching in cavity transmission is limited only by the much weaker – Markovian dephasing or non-radiative decay processes. Our results indicate that particularly for dipolar excitons with enhanced interactions, it should be possible to obtain strong photon blockade effect.

Before proceeding, we note that remarkable progress in realization of photon blockade has been achieved using electromagnetically induced transparency (EIT) [11, 12], which describes the modification of the optical response of a medium stemming from the pumping of a driven atomic or solid-state system into a dark state [17]. Normally, the presence of an excited metastable state that is immune to radiative decay is considered to be an essential requirement for EIT. This metastable state and the ground-state are coupled by coherent laser fields to a common bright state with a large dissipation rate. The dark state that remains uncoupled from the electromagnetic field is a superposition of the ground and metastable states. If the atoms in the metastable state have strong interactions, then it is possible to observe a blockade effect where excitation of a second nearby atom to its metastable state is prohibited. This is the essence of Rydberg blockade where the strong dipolar interactions between atoms lead to quantum correlations between transmitted photons [11].

As we argue below, the cavity-TMD system we are analyzing forms an analog of EIT if we associate the cavity mode with the bright resonance and the exciton mode with the metastable resonance. The Hamiltonian of the system is

$$H = H_{\text{TMD}} + H_{\text{cavity}} + H_{\text{int}} + H_{\text{laser}} + H_{\text{bath}} \quad (1)$$

where

$$H_{\text{TMD}} = \sum_{k_{||}} \left[ \omega_{\text{exc}}(k_{||}) x_{k_{||}}^\dagger x_{k_{||}} + U_{\text{cav-x}} \sum_{k_{||}',q} x_{k_{||}}^\dagger q x_{k_{||}}^\dagger y_{k_{||}'} q x_{k_{||}'} y_{k_{||}} \right] \quad (2)$$

$$H_{\text{cav}} = \omega_c a_c^\dagger a_c \quad (3)$$

$$H_{\text{bath}} = \sum_{k} \omega_k b_k^\dagger b_k + \sum_{k} [ \xi_k a_k^\dagger b_k + \text{h.c.}] \quad (4)$$
\[ H_{\text{int}} = \sum_{k_\parallel} \left[ g_c F^* (k_\parallel) x^\dagger_{k_\parallel} a_c + h.c. \right], \]  
\[ H_{\text{laser}} = [\Omega_0 a_c + h.c.]. \]  

Here, we assumed exciton coupling to a single zero dimensional (0D) fundamental cavity mode \( a_c \) in a structure where the photonic confinement along the \( z \)-direction is much stronger than the lateral confinement. To simplify the expressions, we set \( \hbar = 1 \) and expressed frequencies in a frame rotating with the incident optical frequency \( \omega_L \). As a consequence, \( \omega_{\text{exc}}(k_\parallel) \rightarrow \omega_{\text{exc}}(k_\parallel = 0) - \omega_L + k^2_{||}/(2m_{\text{exc}}) \) and \( \omega_c \rightarrow \omega_c - \omega_L \); here, \( k_\parallel \) denotes the in-plane momentum of the exciton. The exciton-exciton interaction is described as a contact interaction with strength \( U_{x-x} \); this is justified in the low density limit of interest even for dipolar 2D excitons. To describe the coupling between the excitons and the cavity mode, we used the definition \( a_c = \sum_{k_\parallel} F(k_\parallel) a_{k_\parallel} \), where \( F(k_\parallel) \) is the fourier transform of the cavity mode function in the plane of the TMD flake, and \( a_{k_\parallel} \) are the annihilation operators for the 2D cavity field modes of momentum \( k_\parallel \). By integrating out the cavity coupling to free-space vacuum modes \( b_k \) described by \( H_{\text{bath}} \) in Markov approximation, we obtain the Heisenberg equations of motion that includes the cavity decay at rate \( \kappa_c = \xi^2 \rho(\omega_c) \), where \( \rho(\omega_c) \) is the density of states of the free space radiation modes, as well as the associated noise terms. \( \Omega_0 \) is the coupling strength between the coherent probe laser and the cavity field.

The assumed form of exciton-cavity coupling and the absence of direct coupling of excitons to free space vacuum modes is central to our analogy between the cavity-TMD system and the EIT setup. To justify this form, we first recall that due to conservation of in-plane momentum in a translationally invariant 2D cavity-exciton system, each exciton mode with in-plane momentum \( k \) \([13-21]\) couples exclusively to a single 2D cavity mode with identical momentum with strength \( g_c = \sqrt{\Gamma_{\text{rad}} c}/L_z \). Here \( \Gamma_{\text{rad}} \) is the spontaneous emission rate of excitons in free-space, \( c \) is the speed of light and \( L_z \) is the length of the 2D cavity along the direction orthogonal to the monolayer plane. The case where a 0D cavity mode couples to 2D excitons can also be described approximately by \( g_c \), if the in-plane mode confinement is weak such that \( F(k_\parallel) \) can be approximated by a delta function \( \delta_{k_\parallel,0} \). In this regime, the eigenmodes of the coupled system will consist of lower and upper polariton modes that are split by an energy \( \simeq 2g_c \). We emphasize that translationally invariant excitons embedded in a cavity acquire a finite decay rate exclusively due to their coherent coupling to the cavity with a finite mirror loss rate \( \kappa_c \). Moreover, fast excitonic radiative decay \( \Gamma_{\text{rad}} \) in free-space emerges as an advantage, since large \( \Gamma_{\text{rad}} \) leads to enhanced coherent coupling \( g_c \).

The analogy between the cavity-TMD system and the conventional EIT setup employed with Rydberg atoms is a direct consequence of vanishing direct radiative decay of the exciton mode. As indicated in Figure 1 in the cavity-TMD scheme, the role of collective excitation from the ground level to the first excited p-level in Rydberg-EIT is replaced by the cavity mode excitation. The counterpart of the coherent laser coupling of the p-level to the metastable Rydberg state is the vacuum-field coupling of the cavity mode to the TMD exciton. The EIT condition in the Rydberg scheme is achieved by creation of a superposition excitation of the ground and Rydberg states that suppresses light scattering from the intermediate p-level. In the cavity-TMD scheme, the corresponding dark state is a coherent superposition of the ground state with an excitonic excitation with vanishing cavity-mode amplitude

\[ |\Psi\rangle \simeq (\alpha + \beta x^\dagger_0)|0, G\rangle, \] 

where \( |0\rangle \) and \( |G\rangle \) denote the vacuum state of the cavity and the TMD, respectively. Expression in Eq. 7 is the steady-state of the coupled system in the limit of weak drive, provided that the incident drive laser and the bare exciton transition are on resonance \([\omega_{\text{exc}}(k = 0) = 0]\). Upon formation of this coherent superposition, the cavity mode occupancy and consequently cavity transmission vanishes and the incident field experiences perfect reflection. On the other hand, when the drive laser is resonant with the polaritonic transitions in the cavity-TMD system, the transmission spectrum of the system exhibits bright resonance peaks.

Keeping the correspondence with Rydberg blockade, we envision two scenarios: in the first case, we assume \( \omega_{\text{exc}}(k = 0) = \omega_c \) and \( \kappa_c \gg 4g_c \) where the coupled system exhibits no polariton splitting but a dark resonance in transmission. Following the EIT analogy, we find that the width of the transmission dip on resonance is given by \( g_c^2/\kappa_c \). If the TMD excitons are subject to non-radiative decay \( \gamma_{\text{nr}} \) or pure dephasing \( \gamma_{\text{deph}} \) stemming from coupling to Markovian reservoirs, the condition for the observation of a dark resonance is given by \( g_c^2/\kappa_c > \gamma_{\text{deph}} \). Observation of quantum correlations between transmitted photons in this regime would in turn require \( U_{x-x} \gg g_c^2/\kappa_c > \gamma_{\text{deph}} \). This simple analysis indicates that strong photon antibunching is observable even when \( \Gamma_{\text{rad}} \gg U_{x-x} \), provided Eq.(8) is satisfied.

Recent experimental observations suggest that \( \gamma_{\text{deph}} \) plays a relatively minor role in comparison to disorder scattering in determining the non-radiative line broadening of TMD excitons. Therefore, the resonant case that we have just described will in practice be limited by disorder scattering that leads to a strongly energy-dependent, non-Markovian line broadening for low momentum excitons. The non-Markovian nature of the disorder induced decay rate follows from the fact that scattering processes due to disorder conserve energy, and
the phase space available for the exciton to scatter into can be strongly energy dependent. The disorder induced decay rate is determined by the imaginary part of the corresponding self energy \( \Im \{ \Sigma_{\text{dis}}(\omega) \} \) (see Supplementary Material). The frequency window in which \( \Im \{ \Sigma_{\text{dis}}(\omega) \} \) is non-zero is typically of the same order of magnitude as its maximum: we denote the latter as \( \delta_{\text{dis}} \). When \( g_c^2/\kappa_c \leq \delta_{\text{dis}} \), the effects of disorder can be approximated by that of an effective Markovian reservoir. In the opposite limit \( g_c^2/\kappa_c > \delta_{\text{dis}} \), disorder has a vanishing effect on the transmission; the asymmetry of the polariton transmission peaks stemming from quantum interference can be observed in this regime.

The suppression of non-Markovian line broadening in the limit where coherent coupling exceeds Doppler broadening was already highlighted in the context of EIT assisted sum frequency conversion in Doppler broadened atomic gases [23]. Similarly, strongly non-Markovian nature of disorder broadening ensures that the cavity-exciton system exhibits steep dispersion when the dark-resonance width exceeds the disorder broadening. As an important consequence, exciton-exciton interactions can render the system anharmonic even in the limit where interaction strength is weaker than the exciton decay rate. Nevertheless, we find that strong photon antibunching in this limit can only be observed in the limit \( U_{x-x} \geq \max \left\{ \delta_{\text{dis}}, g_c^2/\kappa_c \right\} \).

The overcome this limitation, we consider a second scenario where we assume a large detuning between the cavity and exciton resonances \( [\Delta_e = \omega_c - \omega_{exc}(k = 0) > g_c] \). In this limit, the coupled system exhibits a narrow bright resonance red detuned from the bare exciton resonance by \( g_c^2/\Delta_e \). This case is analogous to Rydberg blockade experiments in the regime where the incident photons are detuned from the intermediate state. However, due to the strong non-Markovian character of the disorder broadening, we find that it is possible to completely suppress the adverse effects \( \Im \{ \Sigma_{\text{dis}}(\omega) \} \). In Fig. 2, we plot the transmission spectrum of the lower polariton for different values of \( g_c \) from 14.5 – 17.5 meV with 1.5 meV intervals, and observe the recovery of transmission at the lower polariton resonance as the detuning of the lower polariton from the bare exciton exceeds \( \delta_{\text{dis}} \) (i.e., \( g_c^2/\Delta_e \gg \delta_{\text{dis}} \)). As a result, we obtain strong photon antibunching even in the limit where \( U_{x-x} < \delta_{\text{dis}}, g_c^2/\kappa_c \), as long as the detuning of the narrow bright resonance is much larger than the strength of the disorder \( g_c^2/\Delta_e > \delta_{\text{dis}} \) and \( U_{x-x} > \max \left\{ g_c^2/\Delta_e, \delta_{\text{dis}} \right\} \). In stark contrast, Markovian processes that lead to the same exciton line broadening as disorder scattering would have lead to Poissonian statistics of the transmitted light [Fig. 3].

The calculate the photon correlation function \( g^{(2)}(t) \), we use the scattering matrix approach presented in Ref. [24], which is reviewed in the Supplementary Material. The conventional wisdom [17] suggests that probe photons injected at an energy where the transmission has the sharpest features result in the largest amplification of the interaction effects in photon correlations. When the probe laser is tuned on resonance with the sharp lower polariton transmission feature, injection of the first photon into the cavity-exciton system will shift the resonance by \( \approx U_{x-x} \). Since the conditional probability that the successive photons will be transmitted (reflected) is reduced (enhanced) in this limit, we expect to see strong photon antibunching (bunching) in \( g^{(2)}(t) \).

For the \( g^{(2)}(t) \) calculation depicted in Fig. 3 we choose \( \Delta_e = 100 \text{ meV}, \ g_c = 20 \text{ meV}, \ \delta_{\text{dis}} = 1 \text{ meV} \) and \( U_{x-x} \approx 10 - 20 \mu eV \). The experimentally reported values of \( g_c \) range from 10 meV to more than 40 meV, depending on the employed cavity structure. Recent experiments demonstrating TMD monolayers as atomically thin mirrors indicate that in clean samples disorder broadening can indeed be as narrow as 0.5 meV and possibly lower. The principal unknown parameter is \( U_{x-x} \); the value we chose was motivated by recently measured interaction strength of GaAs excitons confined to \( A = 2 \mu m^2 \) [25,26]. While a detailed calculation taking into account non-local screening effect [27,28] has not been carried out for \( U_{x-x} \), we expect TMD exciton interaction strength to be comparable to that in GaAs.
To further enhance $U_{x-x}$ it is desirable to use a heterostructure where an intra-layer exciton couples resonantly to an inter-layer (indirect) exciton by coherent electron or hole tunneling ($J$); such structures have been implemented in GaAs structures to realize dipolar polaritons [29] with enhanced interactions [30, 31]. In the limit where the indirect exciton is tuned into resonance with the bright resonance and $g^2_0/\Delta_c > J > \Delta_{dis}$ is satisfied, it would be possible to obtain a bright resonance with a permanent dipole moment.

In the $g^{(2)}(t)$ calculations depicted in Fig. 3 (green and red curves), we take into account radiative decay and disorder scattering, but neglect line broadening of excitons stemming from coupling to additional reservoirs ($\gamma_d$). Long-wavelength phonon coupling between high and low momentum intra-valley excitons, as well as relaxation of bright intra-valley excitons into inter-valley dark exciton states by short-wavelength phonon emission could lead to $\gamma_d > 0$ and limit the minimum achievable linewidth of the bright polariton resonance. We emphasize however, that due to the non-Markovian character of the phonon bath at ultra-low temperatures, strong exciton-cavity coupling could strongly suppress both of these channels; in particular, by choosing $g^2_0/\Delta_c$ to be comparable to the electron-hole exchange interaction, it is possible to eliminate relaxation into dark exciton states by short wavelength phonons. Another potential decay channel for high momentum excitons which will modify the imaginary part of exciton self-energy is radiative coupling to guided modes which in turn have a finite lifetime due to the finite sample size. Coupling to the guided modes result in a frequency independent (Markovian) contribution to the excitonic self energy; the latter decay channel may be suppressed by using in-plane photonic band-gap structures eliminating guided modes that are resonant with the lower-polariton mode.

In summary, we show that photon blockade regime can be achieved in a cavity-TMD system even when the excitation-exciton interaction strength is much smaller than the cavity and exciton radiative decay rates. The resilience of quantum correlations to disorder scattering stems from the non-Markovian nature of the associated exciton coherence decay. Remarkably, the only fundamental requirement for observation of strong photon antibunching is $U_{x-x} > \gamma_d$. Given the immense possibilities for controlling the excitonic properties of TMD monolayers using electrical gates or structured dielectric environment, we expect the demonstration of photon blockade...
to establish cavity-TMD system as a building block of strongly correlated photonic systems.

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