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

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Narrow bright or 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 ultrafast radiative decay rate inside a microcavity, it is possible to obtain extremely narrow dark or bright resonances in transmission. While breaking of translational invariance sets a limit on the width of the dark resonance width, it is possible to obtain a narrow bright resonance that is much narrower than the cavity and bare exciton decay rates and is protected 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 linewidth of the bare exciton. Our findings suggest that atomically thin semiconductors which exhibit large exciton-cavity coupling and small nonradiative line broadening could pave the way for the realization of strongly interacting photonic systems in the solid state.

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Realization of nonperturbative deterministic interactions between single photons remains an outstanding challenge. The necessary conditions for achieving this milestone are (i) strong light-matter (emitter) *coupling* and (ii) strong *interactions* (nonlinearity) between the quanta of matter excitations. While the latter results in quantum correlations between the matter excitations, the former ensures that these correlations are faithfully transferred to propagating light modes that are experimentally accessible.

The figure of merit for the strength of light-matter coupling is cooperativity C, which measures how effectively the emitter decays into a set of photonic modes that can be subsequently detected, as compared to its radiative or nonradiative decay into undetected degrees of freedom. Generically, there are two routes to increasing the cooperativity between an emitter and the light modes. The first option is to manipulate the photonic degree of freedom, either by tightly focusing the light mode onto the emitter [1], or by enhancing the photonic density of states via a cavity [2]. Second, C can be increased in a many-emitter system [3,4] by employing collective states which have a large overlap with the experimentally accessible light modes [5,6]. Excitons embedded in a cavity combine both of these approaches for enhancing C.

On the other hand, the strength of the interactions between matter excitations is generically inversely proportional to the spatial confinement of the excitations, indicating that enhancing C in a many-emitter system typically leads to a reduction in nonlinearity. As a consequence, the observation of quantum correlations between photons generated by delocalized matter excitations not only requires a large C, but also requires that the interaction strength U between the quanta of delocalized polarization waves (polaritons) exceeds their total decay rate Γ_{pol}. It is generally argued that the principal experimental challenge for the realization of a scalable system of strongly interacting polaritons in the solid state is finding systems where U exceeds the cavity and exciton decay rates.

Here, we show theoretically that the collective optical excitations of a two-dimensional (2D) semiconductor crystal embedded in an optical cavity, termed exciton polaritons, can be used to address these challenges. We find that the crucial requirement to achieve this goal is the existence of an exciton resonance with fast radiative decay (Γ_{exc}) that dominates over disorder broadening. We argue that the combination of the large vacuum Rabi splitting that results from fast Γ_{exc} and the non-Markovian nature of disorder scattering allows for the observation of a strong photon blockade effect [7–14] in the solid state. In particular, we show that when the exciton resonance is red detuned from the cavity mode (Fig. 1), the large Rabi splitting can be used to ensure that the lower polariton mode is protected from both the disorder-induced non-Markovian decay and the radiative decay through coupling to the cavity. As a result, the linewidth of the (bright) transmission *peak* associated with the lower polariton mode (i.e., Γ_{pol}) can be much smaller than the exciton disorder broadening as well as the radiative decay rate of the bare exciton and cavity modes. In this limit, the exciton-exciton interaction strength U required to observe large photon antibunching in cavity transmission is limited only by the—much weaker—Markovian dephasing or nonradiative decay processes. Since transition-metal dichalcogenide (TMD) monolayers such as $MoSe_2$ or WSe_2 [15–18] are the only currently available semiconductors that exhibit predominantly radiatively broadened excitonic resonances [19–21], we focus our discussion on this material system; we note that the ultrasmall Bohr radius a_B of TMD excitons implies large cooperativity C.

Before proceeding, we note that remarkable progress in the realization of photon blockade has been achieved using electromagnetically induced transparency (EIT) [9,10], which describes the modification of the optical response of a medium

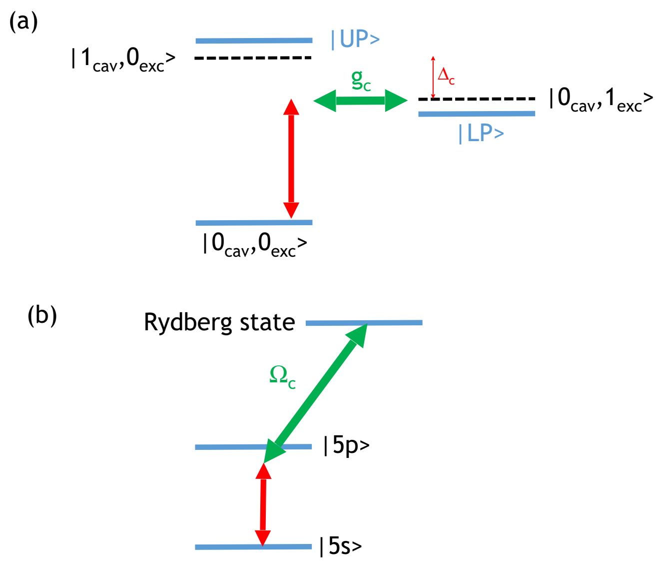

FIG. 1. The summary of the analogy between (a) the cavity-TMD and (b) conventional Rydberg EIT schemes. The role of the fast decaying $5p$ state of the Rydberg atom is fulfilled by the radiatively broadened cavity mode excitation, while the counterpart of the metastable Rydberg state is the excitonic transition of the TMD. Finally, the coherent drive in the Rydberg EIT scheme is replaced by the coherent coupling of the excitonic transition of the TMD to the cavity mode. In this work, we are primarily focusing on the scenario where the cavity mode is blue detuned with respect to the exciton mode by energy Δ_c. In this limit, the coupling at rate g_c between the exciton and the cavity results in two bright transmission resonances: an excitonlike lower polariton (LP) and a cavitylike upper polariton (UP) mode. By choosing $\Delta_c \gg g_c$, the radiative lifetime of the LP $\Gamma_{\rm LP}$ can be prolonged arbitrarily.

stemming from the pumping of a driven atomic system into a dark state [22]. Normally, the presence of an excited metastable state that is immune to radiative decay is considered to be an essential requirement for EIT. 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 the Rydberg blockade [Fig. 1(b)] where the strong dipolar interactions between atoms lead to quantum correlations between transmitted photons [9].

We argue below that the cavity-TMD system we are analyzing forms an analog of EIT where the metastable state has a non-Markovian decay rate induced by disorder, if we associate the cavity mode with the bright resonance and the exciton mode with the metastable resonance. The Hamiltonian of the cavity-TMD system is

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

where

$$H_{\rm TMD} = \sum_{k_\parallel} \omega_{\rm exc}(k_\parallel) x^\dagger_{k_\parallel} x_{k_\parallel} + U_{x-x} \sum_{k_\parallel, k'_\parallel, q} x^\dagger_{k_\parallel+q} x^\dagger_{k'_\parallel-q} x_{k'_\parallel} x_{k_\parallel}, \quad (2)$$

$$H_{\rm cav} = \omega_c a^\dagger_c a_c, \quad (3)$$

$$H_{\rm bath} = \sum_{\mathbf{k}} \omega_{\mathbf{k}} b^\dagger_{\mathbf{k}} b_{\mathbf{k}} + \sum_{\mathbf{k}} [\xi_{\mathbf{k}} a^\dagger_c b_{\mathbf{k}} + {\rm H.c.}], \quad (4)$$

$$H_{\rm int} = \sum_{k_\parallel} [g_c F^*(k_\parallel) x^\dagger_{k_\parallel} a_c + {\rm H.c.}], \quad (5)$$

$$H_{\rm laser} = [\Omega_0 a_c + {\rm H.c.}], \quad (6)$$

$$H_{\rm dis} = \sum_{k_\parallel, k'_\parallel} V_{k_\parallel, k'_\parallel} x^\dagger_{k_\parallel} x_{k'_\parallel}. \quad (7)$$

Here, we assumed that the exciton transition is 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 express frequencies in a frame rotating with the incident optical frequency ω_L. As a consequence, $\omega_{\rm exc}(k_\parallel) \to \omega_{\rm exc}(k_\parallel = 0) - \omega_L + k_\parallel^2/(2m_{\rm exc})$ and $\omega_c \to \omega_c - \omega_L$; 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 $b_{\mathbf{k}}$ described by $H_{\rm bath}$ in the Markov approximation, we obtain the Heisenberg equations of motion that include the cavity decay at rate $\kappa_c \equiv \xi^2 \rho(\omega_c)$, where $\rho(\omega_c)$ is the density of states of the free-space radiation modes, as well as the associated noise terms. Ω_0 is the coupling strength between the coherent probe laser and the cavity field.

$H_{\rm dis}$ describes the processes where the excitons are scattered by disorder. The disorder potential is characterized by the variance of the disorder strength and the disorder correlation length, denoted by σ and η, respectively. The disorder-induced decay rate is determined by the imaginary part of the corresponding self-energy ${\rm Im}[\Sigma_{\rm dis}(\omega)] \equiv \gamma_d(\omega)$. We calculate the disorder-averaged self-energy in the correlated coherent potential approximation (CPA) introduced in Ref. [23] (see Supplemental Material [24]). Correlated CPA allows us to determine the disorder parameters $\frac{\hbar^2\eta^2}{2m_x} = \sigma \approx 1.5$ meV and consequently $\eta \approx 50$ nm by a fit to the experimental data in Ref. [19]. Here, we took the TMD exciton mass m_x to be equal to the bare electron mass m_0. Most importantly for the results discussed below, the correlated CPA approach allows us to fully capture the non-Markovian nature of the disorder-induced decay rate. With these parameters, the energy window in which $\gamma_d(\omega)$ is nonzero is typically of the same order of magnitude as its maximum value $\delta_{\rm dis} = 1$ meV (see inset of Fig. 2). Intuitively, 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 is strongly energy dependent.

The form of exciton-cavity coupling and the absence of direct coupling of excitons to free-space vacuum modes in Eqs. (1)– (7) imply that when the monolayer is embedded in a cavity, the only source of polariton radiative decay is

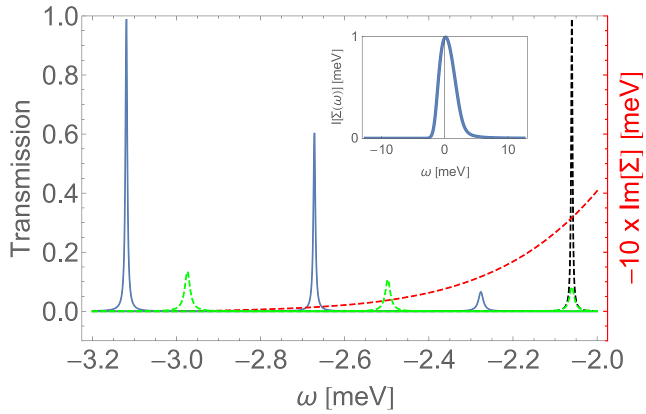

FIG. 2. Transmission ($|t|^2$) spectrum of the lower polariton for $\kappa_c = 0.2$ meV, $\Delta_c = 100$ meV for increasing values of the cavity coupling $g_c = 14.5$–17.5 meV, with 1.5-meV intervals. The energy $\omega_{\rm exc}(0) = 0$ in the rotating frame. The red dashed line is the rescaled low-energy tail of the disorder-induced decay rate $\gamma_d(\omega)$, which is non-Markovian. The blue solid traces take into account of the decay of the $k = 0$ exciton due to disorder analyzed using the correlated coherent potential approximation (CPA), where $\delta_{\rm dis} = 1$ meV and $E_c \equiv \frac{\hbar^2\eta^{-2}}{2m} = \sigma$, where η is the correlation length of the disorder potential and σ is the variance of the disorder potential (see Supplemental Material [24]). The dashed green traces only consider a Markovian dephasing rate $\gamma_{\rm M} = \delta_{\rm dis}/100 = 0.01$ meV, for illustration purposes. Lastly, the black dashed trace is depicted to serve as a reference for the transmission of the lower polariton for $g_c = 14.5$ meV, if neither Markovian dephasing nor disorder-induced decay is taken into account. As the g_c is increased such that the $g_c^2/\Delta_c \gg \delta_{\rm dis}$, the lower polariton resonance is redshifted, and eventually reaches an energy region where the disorder-induced decay rate is negligible. As a result, adverse effects of the disorder-induced decay become negligible, while the effect of Markovian dephasing persists. The peak transmission for the case of Markovian dephasing is given by $|\frac{\Gamma_{\rm LP}}{\Gamma_{\rm LP}+\gamma_{\rm M}}|^2$. The difference in the transition frequency of the exciton with Markovian broadening and non-Markovian broadening is due to the real part of the disorder-induced self-energy (see Supplemental Material [24]). Inset: The imaginary part of the excitonic self-energy due to disorder.

cavity-mirror losses. 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 [25–28] couples exclusively to a single 2D cavity mode with identical momentum with strength $g_c = \sqrt{\Gamma_{\rm rad} c/L_z}$ [29]. Here, $\Gamma_{\rm rad}$ 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 cavity mode confinement is weak such that $F(k_\parallel)$ can be approximated by a delta function $\delta_{k_\parallel,0}$. In this regime, the (coherent) coupling between the $x_{k_\parallel=0}$ and a_c modes results in lower and upper polariton modes that are split by an energy $\simeq 2g_c$, for vanishing cavity-exciton detuning $\Delta_c = \omega_{\rm exc}(k_\parallel = 0) - \omega_c$. The general expression for the radiative decay rate of the lower polariton mode in turn is given by $\Gamma_{\rm LP} = \kappa_c g_c^2/\Delta_c^2$ and is exclusively due to the finite cavity loss rate κ_c.

Having established the model in Eq. (1) we develop an analogy between the cavity-TMD system and the conventional Rydberg-EIT setup. As indicated in Fig. 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 preparing the system in a (dark) superposition 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,

$$|\Psi\rangle \simeq (\alpha + \beta x_0^\dagger)|0, G\rangle, \tag{8}$$

where $|0\rangle$ and $|G\rangle$ denote the vacuum state of the cavity and the TMD, respectively. The expression in Eq. (8) 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 [$\omega_{\rm exc}(k = 0) = 0$]. Upon the 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 exhibits bright resonance peaks.

Keeping the correspondence with the Rydberg blockade, we envision two scenarios: In the first case, we assume $\omega_{\rm exc}(k = 0) = \omega_c$ and $\kappa_c > 4g_c$ 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/κ_c. However, the TMD excitons can be subject to Markovian or non-Markovian nonradiative decay, whose rates are denoted by $\gamma_{\rm M}$ and $\gamma_d(\omega)$, respectively. In this case, the condition for the observation of a dark resonance is given by $g_c^2/\kappa_c > \gamma_{\rm T}(k = 0) = \gamma_d[\omega_{\rm exc}(0)] + \gamma_{\rm M}$. The observation of quantum correlations between transmitted photons in this regime would in turn require $U_{x-x} \simeq g_0^2/\kappa_c > \gamma_{\rm T}(0)$. This simple analysis indicates that strong photon antibunching is in principle observable even when $\Gamma_{\rm rad} \gg U_{x-x}$.

When $g_c^2/\kappa_c \leqslant \delta_{\rm dis}$, the effects of disorder can be approximated by that of an effective Markovian reservoir. In the opposite limit $g_c^2/\kappa_c > \delta_{\rm dis}$, disorder has a vanishing effect on the transmission and exciton-exciton interactions can render the system anharmonic even in the aforementioned limit $\Gamma_{\rm rad} \gg U_{x-x}$. However, we find that strong photon antibunching in this limit can only be observed only if $U_{x-x} \geqslant \max[\delta_{\rm dis}, g_c^2/\kappa_c]$.

To overcome this limitation, we consider a second scenario where we assume a large detuning between the cavity and exciton resonances [$\Delta_c = \omega_c - \omega_{\rm exc}(k = 0) > g_c$]. In this limit, the coupled system exhibits a narrow bright resonance red detuned from the bare exciton resonance by g_c^2/Δ_c [Fig. 1(a)]. This case is analogous to Rydberg blockade experiments in the regime where the incident photons are detuned from the intermediate state. Due to the strong non-Markovian character

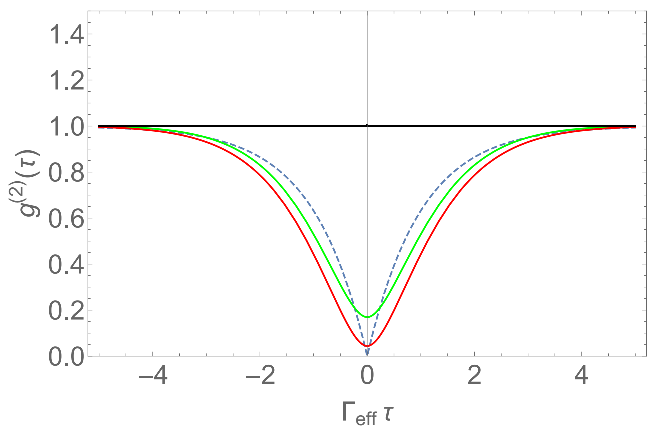

FIG. 3. Time-dependent second-order correlation function $g^{(2)}(\tau)$ when the incoming photons are resonant with the lower polariton in the case of CPA disorder (in red and green) and Markovian dephasing (in black). $U = 0.02$ meV (in red) $U = 0.01$ meV (in green), $g_c = 20$ meV, $\kappa_c = 0.1$ meV, $\Delta_c = 100$ meV. Disorder self-energy is calculated such that $\delta_{\text{dis}} = 1$ meV, and $E_c \equiv \frac{\hbar^2\eta^{-2}}{2m} = \sigma$, where η is the correlation length of the disorder potential and σ is the variance of the disorder potential (see Supplemental Material [24]). For these parameters and interaction strength $U = 0.02$ meV, the Markovian dephasing with $\gamma_{\text{M}} = \delta_{\text{dis}}$ completely destroys the transmission peak of the lower polariton and results in a flat $g^{(2)}(0) \approx 1$, implying Poissonian statistics. The blue dashed line is to guide the eye and represents the exponential decay with the radiative decay rate (Γ_{LP}) of the lower polariton.

of the disorder broadening, we find that it is possible to completely suppress the adverse effects of $\gamma_d(\omega)$.

We demonstrate this effect in Fig. 2, by plotting the transmission spectrum of the lower polariton for different values of g_c from 14.5 to 17.5 meV with 1.5-meV intervals, as well as the rescaled low-energy tail of the non-Markovian decay rate induced by disorder. We observe that the transmission at the lower polariton resonance is recovered as the detuning of the lower polariton from the bare exciton exceeds δ_{dis} (i.e., $g_c^2/\Delta_c > \delta_{\text{dis}}$). We emphasize that the spectrum depicted in Fig. 2 is calculated using the imaginary part of the full cavity Green's function, which incorporates coupling between the cavity field and the exciton, as well as the complete frequency dependence of the excitonic disorder self-energy. As a result, we expect to obtain strong photon antibunching even in the limit where $U_{x-x} < \delta_{\text{dis}}, g_c^2/\kappa_c$, as long as the exciton-exciton interaction strength is larger than the radiative width Γ_{LP} of the lower polariton $[U_{x-x} > \max(\Gamma_{\text{LP}} = \kappa_c g_c^2/\Delta_c^2, \gamma_d)]$ (Fig. 3). In stark contrast, Markovian processes that lead to a much smaller exciton decay rate $\gamma_{\text{M}} = 0.01$ meV substantially diminish the transmission peak (dashed green lines in Fig. 2). The value of γ_{M} is set to demonstrate the contrast between the adverse effects of Markovian and non-Markovian processes. We discuss the possible sources of γ_{M} in the conclusion.

The calculate the photon correlation function $g^{(2)}(\tau)$, we use the scattering matrix approach presented in Ref. [30], which allows us to calculate the effects due to the non-Markovian nature of the disorder-induced decay rate. We review the scattering matrix formalism in the Supplemental Material [24]. Conventional wisdom [22] 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 $g^{(2)}(\tau)$ of the transmitted (reflected) light.

For the $g^{(2)}(\tau)$ calculation depicted in Fig. 3 we choose $\Delta_c = 100$ meV, $g_c = 20$ meV, $\delta_{\text{dis}} = 1$ meV, $\frac{\hbar^2\eta^2}{2m_x} = \sigma$, and $U_{x-x} \simeq 10$–$20\ \mu$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 [19–21] 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 the recently measured interaction strength of GaAs excitons confined to $A = 2\ \mu\text{m}^2$ [31,32]. While a detailed calculation taking into account nonlocal screening effect [33,34] has not been carried out for U_{x-x}, we expect the TMD exciton interaction strength to be comparable to that in GaAs.

To further enhance U_{x-x} it is desirable to use a heterobilayer structure where an intralayer exciton couples resonantly to an interlayer (indirect) exciton by coherent electron or hole tunneling (J); such structures have been implemented in GaAs structures to realize dipolar polaritons [35] with enhanced interactions [36,37]. In the limit where the indirect exciton is tuned into resonance with the bright resonance and $g_c^2/\Delta_c > J > \Delta_{\text{dis}}$ is satisfied, it would be possible to obtain a bright resonance with a permanent dipole moment.

In the $g^{(2)}(\tau)$ calculations depicted in Fig. 2 (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 (γ_{M}). Long-wavelength phonon coupling between high- and low-momentum intravalley excitons, as well as relaxation of bright intravalley excitons into intervalley dark exciton states by short-wavelength phonon emission could lead to $\gamma_M > 0$ 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 ultralow temperatures, strong exciton-cavity coupling could strongly suppress both of these channels; in particular, by choosing g_c^2/Δ_c to be comparable to the electron-hole exchange interaction, it may be possible to eliminate relaxation into dark exciton states by short-wavelength phonons. Another potential Markovian exciton decay channel is disorder-mediated coupling of the LP to guided mode polaritons [38,39]. However, as long as g_c^2/Δ_c is larger than the longitudinal-transverse (LT) splitting of the guided modes of the TMD monolayer, the decay rate induced by coupling to the guided modes is strongly reduced due to the small exciton fraction and lightlike dispersion of the guided mode polaritons [38,39]. Moreover, this 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 a photon blockade regime can be achieved in a cavity-TMD system even when the exciton-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 the observation of strong photon antibunching is $U_{x-x} > \gamma_{\mathrm{M}}$. Given the immense possibilities for controlling the excitonic properties of TMD monolayers using electrical gates or a structured dielectric environment, we expect the demonstration of the photon blockade to establish the cavity-TMD system as a building block of strongly correlated photonic systems [20,21].

Note added. Recently, we became aware of a related article proposing a complementary approach to reach the photon blockade regime [40].

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