Optical non-linearities and translational symmetry breaking in driven-dissipative moiré exciton-polaritons

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(Dated: June 14, 2022)

Moiré lattices formed from semiconductor bilayers host tightly localised excitons that can simultaneously couple strongly to light and possess large electric dipole moments. This facilitates the realization of new forms of polaritons that are very strongly interacting and that have been predicted to lead to strong optical non-linearities controlled by multi-photon resonances. Here, we investigate the role of the non-local component of the exciton-exciton (dipolar) interactions on the optical response of these strongly-interacting moiré exciton-polaritons under conditions of strong optical driving. We find that the non-local interactions can strongly influence the steady-state properties leading to multi-stabilities with broken translational symmetry and pronounced distortions of the multi-photon resonances. We develop a self-consistent approach to describe the steady-state solution of moiré excitons coupled to a cavity field, treating the long-range interaction between the excitons and the photon field at the semi-classical level.

I. INTRODUCTION

In van der Waals bilayers, the moiré superlattice resulting from lattice mismatch or relative twist angle has emerged as a productive means to realize complex quantum many-body phases \cite{1}. The quantum confinement provided by the moiré landscape has unfolded many opportunities towards the controlled realization of strongly correlated electronic phases such as Wigner crystals \cite{2}, Mott insulators \cite{3}, superconductivity \cite{4,5} and more \cite{6–11}. In semiconductor bilayers, moiré materials have unveiled a new class of excitations \cite{12–16}: moiré excitons. Moiré excitons possess properties that make them ideal to explore strongly interacting phases of bosonic matter in uncharted territory, which include prospects for high-temperature and long-lived Bose-Einstein condensates \cite{17,18}, the superfluid-Mott transition \cite{19}, excitonic insulators \cite{20}, and supersolidity \cite{21}.

When combined with an optical cavity, the underlying nature of moiré excitons leads to novel forms of exciton-polaritons and presents new opportunities to engineer hybrid quantum states of light and matter with no equivalent in conventional polaritons. The strong confinement of excitons to the moiré sites yields distinctive moiré-induced polaritons \cite{22}, novel forms of quantum emitters \cite{23–25}, and promises a new generation of polaritons with tuneable features \cite{26,27}. The moiré superlattice activates a rich interplay between the tight confinement of the excitons, the light-matter coupling, and the strong exciton-exciton interactions.

Moiré polaritons are particularly interesting as the underlying excitons can inherit properties of both spatially direct and indirect excitons, which can provide them with valuable features such as sizeable light-matter coupling \cite{12,28} and strong exciton-exciton interactions \cite{22}. Recent experimental \cite{22} and theoretical studies \cite{25} have demonstrated that moiré polaritons feature optical properties with large non-linearities very different from conventional polaritons in semi-conductors. Theoretically, it has been shown that the tight confinement of the excitons to the moiré sites leads to pronounced multi-photon resonances governed by the underlying discrete excitonic energy spectrum \cite{25}, arising as a consequence of the quasi zero-dimensional character of the excitons and their strong local interactions. This is predicted to permit lasing based on single- and multi-photon processes induced by the moiré lattice.

In addition to the strong on-site exciton-exciton interactions, the indirect character of the moiré excitons leads to non-local interactions. While for moiré lattices the on-site interaction is expected to dominate over the non-local interactions, the precise role of the non-local interactions remains relatively unexplored.

Here, motivated by this open question, we study the many-body optical properties of a van der Waals heterostructure bilayer, focusing on the effects of the non-local exciton-exciton interactions arising from their dipolar character. We show that non-local interactions can strongly modify the optical response of the system and demonstrate the emergence of steady states with broken translational symmetry. In addition, the presence of non-local interactions influences the multi-photon resonance conditions leading to a rich phase-diagram with strongly hysteretic features.

The outline of the paper is as follows. In Section II, we detail the model we study – a tight binding model of excitons coupled to cavity photons – and the methods we employ to determine its properties. Here, we introduce three coloured excitonic sites which we treat independently at the mean-field level, we also discuss the mean-field and semi-classical treatment for the cavity photons. In Section III we turn our attention to the study of hard-
core excitons, and we reveal the emergence of steady states with broken translational symmetry that can be accessed through several hysteresis mechanisms. The interplay between the on-site and non-local interactions is unraveled in Section IV where we analyze the effects of the dipolar interactions on the multi-photon resonances. Finally, in Section V we discuss the experimental consequences and outlook based on our results.

II. MODEL AND METHODS

We consider moiré excitons in a van der Waals heterostructure bilayer coupled to a microcavity in the presence of a coherent drive of photons. The moiré landscape leads to flat mini bands that arise from the tight localization of the excitons to the moiré sites. Hence, we describe the excitons via a tight binding Hamiltonian given by

$$\hat{H}_X = \sum_i \omega_X \hat{x}_i^\dagger \hat{x}_i + \frac{U_X}{2} \sum_i \hat{x}_i^\dagger \hat{x}_i \hat{x}_i^\dagger \hat{x}_i + \sum_{i \neq j} V_{ij} \hat{x}_i^\dagger \hat{x}_j \hat{x}_j^\dagger \hat{x}_i,$$

(1)

here $\hat{x}_i^\dagger$ creates an exciton with energy $\omega_X$ in the site $i$, with the sites arranged on a triangular lattice. (We set $\hbar = 1$ throughout.) We neglect the hopping of excitons between local sites – i.e. the bandwidth of the lowest energy exciton band in the moiré lattice. For a wide range of parameters this can be small compared to the transport via the cavity mode. The on-site exciton-exciton interaction is denoted by $U_X$, while $V_{ij}$ corresponds to the interaction between an exciton in site $i$ and an exciton in site $j$. In general, the moiré potential supports multiple localised exciton states [14, 27]. Here we restrict our study to the lowest excitonic state and assume that the energy separation between the first and second bands remains larger than any other typical energy of the system.

The bilayer is embedded in a high-finesse microcavity with the ideal dispersion of the cavity photons described by

$$\hat{H}_1 = \sum_k \omega_c(k) \hat{a}_k^\dagger \hat{a}_k.$$

(2)

Here the free dispersion of photon is $\omega_c(k) = \omega_c + |k|^2/(2m_c)$, where $m_c$ is the cavity photon mass. The operator $\hat{a}_k^\dagger$ creates a cavity photon with in-plane momentum $k$. The coupling between excitons and cavity photons is given by the usual light-matter Hamiltonian

$$\hat{H}_{1-m} = \sum_k \Omega \left( \hat{a}_k^\dagger \hat{x}_k + \hat{x}_k^\dagger \hat{a}_k \right),$$

(3)

where $\hat{x}_k^\dagger$ creates an exciton with in-plane momentum $k$. The strength of the light-matter coupling, denoted by the Rabi frequency $\Omega$, is assumed much smaller than the typical energy of the excitons, so the light-matter Hamiltonian is written under the rotating wave approximation. (For typical systems $\Omega$ is a several meV while $\omega_X$ is on the order of 1 eV.)

To make further progress we shall restrict the light-matter coupling to the $k = 0$ cavity mode [25]. Thus, we simplify the light and light-matter terms of the Hamiltonian as

$$\hat{H}_1 + \hat{H}_{1-m} = \omega_c \hat{a}_0^\dagger \hat{a}_0 + \frac{1}{\sqrt{N}} \sum_i \Omega \left( \hat{a}_0^\dagger \hat{x}_i + \hat{x}_i^\dagger \hat{a}_0 \right),$$

(4)

where $\hat{a}_0$ now refers to the $k = 0$ cavity mode. This approximation assumes a spatially uniform coupling between the cavity photons and excitons occupying different sites in the moiré superlattice. This assumption is justified for two reasons. Firstly, due to the ultra-light mass of the cavity mode, the cavity photons decouple from the excitons when the kinetic energy of the photon becomes of the order of the Rabi coupling $k^2/2m_c = \Omega$. This means that excitons only couple to cavity modes with wavelengths that are larger than a length scale $\lambda = 2\pi/k$, which is typically very large, covering hundreds of moiré sites. This large lengthscale justifies a mean-field treatment of the cavity-mediated exciton-exciton coupling. Secondly, we consider situations in which the cavity mode is pumped uniformly. Specifically, we consider an external coherent injection of photons via

$$\hat{H}_{\text{drive}} = (F \hat{a}_0^\dagger e^{-i\omega_p t} + F^* \hat{a}_0 e^{i\omega_p t}),$$

(5)

where $F$ and $\omega_p$ are the strength and the frequency of the driving term respectively. As will be discussed below, we will employ a mean-field approach for the cavity photons, where excitons couple only to the $k = 0$ cavity mode.

We mention that the ability to create arbitrary number of excitons is limited by the intrinsic nature of the excitons, where its non-bosonic nature leads to saturation effects. Although this remains as an open question [29, 30], such effects can be accounted to a first approximation.
through an anharmonic light-matter coupling term [31–33]. This term, however, only quantitatively modifies the optical properties for small exciton numbers [25].

We allow the system to be lossy, and study the density operator of the system, \( \hat{\rho} \), via the Gorini–Kossakowski–Sudarshan–Lindblad master equation \[ \frac{d\hat{\rho}}{dt} = -i[\hat{H}, \hat{\rho}] + D[\hat{\rho}] = \mathcal{L}[\hat{\rho}], \] (6)

where the total Hamiltonian is given by \( \hat{H} = \hat{H}_X + \hat{H}_l + \hat{H}_{1-m} + \hat{H}_{\text{drive}} \). The dissipative character of the system is accounted for by the operator \[ D[\hat{\rho}] = \frac{\gamma_x}{2} \left[ 2\hat{\rho}\hat{a}^\dagger - \{\hat{a}^\dagger \hat{a}, \hat{\rho}\} \right] + \sum_i \frac{\gamma_x}{2} \left[ 2\hat{x}_i\hat{\rho}\hat{x}_i^\dagger - \{\hat{x}_i^\dagger \hat{x}_i, \hat{\rho}\} \right], \] (7)

here \( \gamma_x \) and \( \gamma_c \) are the damping rate of the excitons and photons respectively.

The spatial stacking of the monolayers leads to a moiré periodicity that confines the excitons into a triangular lattice. To allow spatial ordering of the excitons we introduce a 3-site supercell as illustrated in Fig. 1. We thus define three kinds of sites within a larger unit cell, denoted in the figure (and referred to below) by three different colours: red, green, and blue. By treating these three sites independently, our present study extends beyond Ref. [25] to explicitly permit states with broken translational symmetry. We will show that this can arise as a consequence of the non-local interaction \( V_{nn} \).

We employ a mean-field approximation to decouple the dipolar interactions in Eq. 1. We define a supercell as illustrated in Fig. 1, treating the occupations of all sites of the same colour to be the same. We thus replace \( x_i \rightarrow \hat{x}_\alpha \) where \( \alpha \in \{R,G,B\} \) labels the three distinct sites within the supercell. The Hamiltonian for each colour is then

\[ \hat{H}_{X,\alpha} = \left( \omega_X + V_{od}n_x + V_{od} \sum_{\beta \neq \alpha} n_\beta \right) \hat{x}_\alpha^\dagger \hat{x}_\alpha + \frac{U_X}{2} \hat{x}_\alpha^\dagger \hat{x}_\alpha \hat{x}_\alpha \hat{x}_\alpha, \] (8)

this local Hamiltonian treats the local terms in Eq. 1 exactly, but takes the long-range dipolar interaction at the mean-field level, with the expectation values of the exciton number for the three different sites, \( n_\beta \), to be determined self-consistently. Here, \( V_{od} = 2.124 \times V_{nn} \) and \( V_{od} = 4.455 \times V_{nn} \) give the mean-field dipole-dipole interaction between the site \( \alpha \) in the supercell and all of the different sites with same and different colour respectively, see further details in the Appendix A.

On the other hand, the light-matter coupling is insensitive to our artificial distinction of sites, that is, cavity photons couple equally to excitons regardless of the colour of the site they occupy. The light-matter coupling for a given colour simply reads as

\[ \hat{H}_{1-m,\alpha} = \frac{\Omega}{\sqrt{N}} (\hat{a}^\dagger \hat{x}_\alpha + \hat{a} \hat{x}_\alpha^\dagger). \]

To make further progress, we take a semi-classical approach for the cavity photons, where we replace the cavity field by its expectation value \( \langle \hat{a} \rangle = \sqrt{N} \psi \). In this case, the steady-state solution for the photon amplitude \( \psi \) is given by

\[ \psi = \frac{1}{\Delta_c} \left( f + \frac{\Omega}{3} \sum_{\alpha=R,G,B} \langle \hat{x}_\alpha \rangle \right), \]

(10)

where \( f = F/N \). The last term inside of the brackets accounts for the possible different expectation value of sites with different colour. Here, \( \Delta_c = \Delta_c + i\gamma_c/2 \) with \( \Delta_c = \omega_p - \omega_c \), the detuning of the pump frequency from the cavity.

These approximations permit us to define three Hamiltonians for differently colours \( \alpha \),

\[ \hat{H}_{\text{loc,}\alpha} = \hat{H}_{X,\alpha} + \hat{H}_{1-m,\alpha}, \]

(11)

with \( \alpha, \beta \in \{R,G,B\} \), that are local. The coloured Hamiltonians are coupled through the exciton-exciton interactions and the light-matter coupling. The exciton-exciton interaction couples sites with different and same colours via the terms \( V_{od} \) and \( V_{dd} \). We will treat the supercell level. The light-matter coupling introduces a long-range mediated tunneling, where an exciton in a given site can convert to a cavity photon, which can decay into an exciton in any other moiré site. Thus, the long-range photon-mediated hopping couples sites with the same and different colours. This leads to a coupling between the coloured local Hamiltonians. Our approach assumes that the cavity field retains the spatially uniform coupling to the excitons in the presence of non-local interactions and that the cavity field maintains population only in the \( k = 0 \) mode[25].

Our approach leads to a set of three coupled master equations, for the sites of each different colour

\[ \frac{d\hat{\rho}_\alpha}{dt} = \mathcal{L}[\hat{\rho}_\alpha] \]

(12)

To obtain the steady-state properties we employ exact diagonalization of each of these three equations. They are coupled since, via Eqn. (10), the expectation values of the photon amplitude \( \psi \) and the exciton number \( \langle \hat{x}_\alpha \rangle \) must be obtained self-consistently. The numerical scheme is detailed in the Appendix B.

The long-range interaction between the excitons stems from their indirect nature that leads to a dipole-dipole interaction that scales with separation \( r \) as \( 1/r^3 \). Due to the large moiré periodicity, the on-site interaction \( U_X \) is largely dominant with respect to \( V_{nn} \). While the on-site interaction can be made of the order of some tens.
meV, the interaction between first neighbours is estimated $V_{nn} \sim 0.1 - 1$ meV, thus, for typical experiments one expects $V_{nn}/U_X \approx 10^{-1} - 10^{-2}$, which enables the study of the imprints of the non-local over a wide range of parameters. In view of these estimates, in this work we consider the non-local terms to remain small with respect to the on-site interaction $V_d/U_X \ll 1$ and $V_{od}/U_X \ll 1$. We consider two cases, first, we consider hard-core excitons which are prevented from double occupation. Then, we study the interplay between multi-photon resonances and non-local interactions.

III. HARD-CORE EXCITONS

For clarity, we start our study in the limit of hard-core excitons which explicitly forbids multiple occupation, this case will allow us to understand the effects of the long-range interaction disentangled from the on-site interaction.

For hard-core excitons $U_X \to \infty$ non-linearities arise from the impossibility to create multiple excitons per site and from the non-local interactions. First, we explore steady-state solutions with equal population on the three sites. We start by considering an initial seed for our self-consistent scheme that is population balanced ($n_R = n_G = n_B$), see Appendix B. In this case, we find a pair of solutions corresponding to low- and high-density hysteresis branches. The former corresponds to varying $f/V_{nn}$ from below while the latter arises from $f/V_{nn}$ being tuned from above. These solutions are illustrated in Fig. 2 for $\Omega/V_{nn} = 0.5$, while fixing the losses $\gamma_x/V_{nn} = \gamma_c/V_{nn} = 0.1$ and cavity and exciton detunings of $\Delta\omega_c/V_{nn} = -1$ and $\Delta\omega_x/V_{nn} = 0.9$, respectively. Here, $\Delta\omega_x = \omega_p - \omega_X$, is the detuning of the exciton energy from the pump frequency.

![Fig. 2. Exciton number per site for the low- and high-density branches of the hysteretic response, in the hard-core limit $U_X \to \infty$. Here we consider only uniform states in which $n_\alpha$ is independent of site $\alpha$. We take $\Omega/V_{nn} = 0.5$, with $\gamma_x/V_{nn} = \gamma_c/V_{nn} = 0.1$, and the detunings are $\Delta\omega_c/V_{nn} = -1$ and $\Delta\omega_x/V_{nn} = 0.9$.](image)

That strong on-site interactions and the inherent nature of moiré exciton-polaritons can give rise to new physical phenomena absent in conventional polaritons [22, 25]. Fig. 2 illustrates that the transition from a low-density regime ($f/V_{nn} \lesssim 0.05$) to a regime dominated by the driving ($f/V_{nn} \gtrsim 0.3$) is separated by an intermediate regime where exciton-exciton interactions lead to bistability. Intriguingly, as we now show, in this regime the presence of non-local interactions can lead to additional features, including steady-state solutions with colour-unbalanced populations, that is, with spatial ordering of the excitons. These solutions are illustrated in Fig. 3 for the same values of the parameters as in Fig. 2.

![Fig. 3. Exciton number per site. (a) Steady-state solutions with $n_B = n_G > n_R$, i.e. one site of low density. (b) Steady-state solutions exhibiting a regime with $n_R > n_B = n_G$, i.e. one site of high density. The pink area illustrates the regime with unbalanced populations. Parameters are the same as for Fig. 2.](image)

The ranges of values of the drive corresponding to states with broken translational symmetry are shaded in pink. In these regimes the different sites (labelled by different colours) have unbalanced populations.

Figure 3(a) shows steady-state solutions obtained via hysteresis where a regime with broken translational symmetry emerges, characterised by $n_B = n_G \neq n_R$. To reach these steady states, the initial seed of our self-consistent scheme is required to have the same broken translational symmetry as the final state. Therefore, we consider an initial seed that retains hysteretically the solutions for the blue and green sites while sets to zero the density and coherence of the red sites, see further details in Appendix B. In this regime (pink area), two solutions remain in a high-density phase below the bifurcation point, whereas the third colour slowly transits to low densities. Figure. 3(a) illustrates the case where blue and green moiré sites are in a high density phase and the red tends to low densities. However, one can find the equivalent solutions with $n_B = n_R \neq n_G$ and $n_R = n_G \neq n_B$ that exhibit identical features (not shown).

Besides, additional solutions arise where only one coloured site suddenly jumps into a high-density phase at expenses of two moiré sites less populated. These solutions are illustrated in Fig. 3(b) where the red sites prevail in a high-density phases with much larger popu-
lation than the blue and green exciton sites, here \( n_R \neq n_B = n_G \). These steady-state solutions are accessed with a seed where the initial self-consistent parameters for the blue and green sites are set to zero while for the red sites the high-density hysteresis branch is followed, see details in Appendix B. We have explored different self-consistent schemes, but have not found fully population-imbalanced metastable states, that is, with \( n_R \neq n_B, n_B \neq n_G \), and \( n_R \neq n_G \).

The photon amplitude in Eq. 10, which couples collectively to the sites with different colours, can also be used as a witness of this set of solutions with broken translational symmetry for the excitons. This is illustrated in Fig. 4 where we show the photon number \( n_p \sim |\psi|_A|^2 \) for the various hysteresis branches discussed above. Since for excitons the low to high density transition occurs for different values of \( f \), the photon number also exhibits this strong hysteresis dependence. Figure 4(b) shows the photon number for the same parameters as Figs. 2-3. Here, the black lines correspond to the photon densities when the excitons are uniformly distributed in the moiré lattice, that is, the population-balanced low- and high-density branches shown in Fig. 2. The red lines illustrate the photon densities of steady-state solutions for which the exciton occupations exhibit broken translational symmetry as in Fig. 3(a)-(b). The pink area corresponds to the regime where a steady state can be found with broken translational symmetry.

These imbalanced solutions are sensitive to the ratio \( \Omega/V_{nn} \) and disappear when this parameter is large enough. In Fig. 4 we show the shrinking of these solutions when \( \Omega/V_{nn} \) is increased, from \( \Omega/V_{nn} = 0.35 \) to 0.5. The pink region clearly decreases for larger values of \( \Omega/V_{nn} \). This illustrates that the solutions with broken translational symmetry are suppressed as for large \( \Omega/V_{nn} \), and fade for \( \Omega/V_{nn} > 0.65 \). The broadening of the excitonic lines leads to smoothing of the interaction effects, in turn, the large moiré periodicity which gives a small \( V_{nn} \) imposes narrow excitonic lines. Experimentally, small broadening of the exciton linewidths have been reported in moiré setups, which can be of the order of 0.1meV [13].

Thus we have shown that non-local interactions allow for steady-state solutions with broken translational symmetry, that is, with unbalanced population in terms of the exciton colour. Numerically, we access states with broken translational symmetry by initializing our self-consisting approach with seeds that explicitly break this symmetry, see further details in the Appendix B. Our ansatz is based on the uses of a supercell containing three kinds of moiré sites, thus, the solutions follow the restriction imposed by our ansatz. In general, one would expect that in presence of disorder different solutions beyond the limitations of our ansatz can emerge giving the solutions a more complex spatial structure.

**IV. MULTI-PHOTON RESONANCES AND NON-LOCAL INTERACTIONS**

Now, we turn our attention to the study of the interplay between the on-site interactions and the non-local interactions. Thus, we relax the hard-core constraint and allow for multiple occupation. In absence of non-local interactions, that is, for \( V_{nn} = 0 \), the phase-diagram is governed by the multi-photon resonance condition

\[
N\omega_p = N\omega_X + \frac{U_X}{2}N(N-1),
\]

which leads to the condition

\[
\frac{2\Delta\omega_X}{U_X} = (N-1).
\]

Physically this can be understood in terms of an energetic condition dictating that \( N \) exciton resonances are promoted whenever the energy of \( N \) non-interacting photons matches the energy of \( N \) interacting excitons [25]. In

**FIG. 4.** Photon amplitude for the several hysteresis mechanism. (a) \( n_p \) for \( \Omega/V_{nn} = 0.35 \) and (b) \( \Omega/V_{nn} = 0.5 \) and remaining parameters as in Figs. 2 and 3. Black lines correspond to steady-state solutions with population balanced exciton number. The photon numbers for steady states with spatial ordering of excitons are illustrated by the red curves. The pink area illustrates the regime where excitons can have density order.

**FIG. 5.** Exciton number per site for steady states with balanced colour population. (a) Low-density hysteresis branch and (b) high-density hysteresis branch. We take \( V_{ad}/U_X \approx 0.1 \), \( \Omega/U_X = 0.65 \), \( \Delta\omega_c/U_X = -1 \) and \( \gamma_c/U_X = 0.2 \).

In the presence of non-local interactions, we expect a shift of
this energetic condition: treating $V_{nn}$ at the mean-field level, the interaction between adjacent excitons simply displaces the on-site energy

$$\omega' = \omega + \left(V_{d}n_{\alpha} + V_{od} \sum_{\beta \neq \alpha} n_{\beta}\right),$$

thus, physically, one anticipates that for a site with a given colour $\alpha$, the resonance is displaced to

$$N\omega_{p} = N \left(\omega + V_{d}n_{\alpha} + V_{od} \sum_{\beta \neq \alpha} n_{\beta}\right) + \frac{U_{X}}{2} N(N - 1).$$

(15)

In this case, the multi-photon resonance (14) depends on $n_{\beta}$ and is not longer necessarily an integer. From our earlier analysis on the effects of the non-local interactions we also anticipate the emergence of multi-stabilities. Note that, although the cavity field and $V_{nn}$ are treated at the mean-field level, we still perform a full quantum calculation for the driven excitons on a single site.

We begin by discussing the case of large ratio $\Omega/V_{nn}$, which as explained above, tends to inhibit solutions with spatial ordering. We take $\Omega/U_{X} = 0.65$ which corresponds to $\Omega/V_{nn} = 1.42$, finally, we consider $V_{od}/U_{X} = 0.1$. In Fig. 5 we show the solutions found for a finite on-site interaction. We turn our attention first to population-balanced solutions obtained with initial seeds that do not break translational symmetry ($n_{R} = n_{B} = n_{G}$) and find two solutions corresponding to the low-density and high-density hysteresis branches.

Figure. 5(a) corresponds to the phase-diagram following the low-density branch, that is, $f/U_{X}$ being tuned from below. In this case, the phase-diagram shows sharp cusp-like features at detunings closely governed by the bare multi-photon resonance in Eqs. 13, 14. Figure. 5(a) corresponds to the case where the low to high density transition is promoted from below, that is, it corresponds to $n_{\beta} \approx 0$ in Eq. 15, and therefore the cusp-like features are barely shifted away from the resonance condition Eq. 13. The non-local interaction does introduce some blurring of the cusps, but the locations remain closely tied to Eq. 14.

On the high-density hysteresis branch, on the other hand, the transition is crossed from above. In this case, the exciton number of the adjacent sites is relatively large $n_{\beta} \neq 0$, hence, the multi-photon resonances in Eq. 15 acquire large energy shifts away from Eq. 14 and visibly distort the phase-diagram. The deviations of the multi-photon resonances and the profound hysteresis contrast with the case in Ref. [25] where the high-density hysteresis branch respects the position of the multi-photon resonances. Therefore, one of the measurable consequences of the non-local interactions are shifts and broadenings of the lobular pattern. In the regime where the non-local interactions are further suppressed with respect to the on-site interactions, for instance $V_{od}/U_{X} \approx 0.05$ one obtains a phase-diagram that closely follows the bare and discrete multi-photon resonances [25], as shown explicitly in Fig. 9 of Appendix. C. Importantly, by means of the twist angle, one can therefore, enhance or suppress the effects of the non-local interactions on the optical response of the system [21].

We also find that the non-local interactions lead to more metastable states, even within the space of population-balanced solutions. This multistability is somewhat reminiscent of the multistability seen in Fig. 3 and Fig. 5 and consists of steady state solutions with modified multi-photon resonance patterns. However we emphasise that it differs from the cases presented in Section. III in that the populations remain balanced. A set of four metastable solutions can be accessed through different hysteresis schemes which retain balanced populations, see Appendix C.

For finite $U_{X}$ one can also obtain states with broken translational symmetry, such solutions require, however, smaller values of $\Omega/V_{nn}$. In Fig. 6 we show, for $\Omega/V_{nn} = 0.45$, solutions with unbalanced populations where red coloured exciton sites jump to a high density state with blue and green excitons smoothly increasing their density. One recognizes the similarities between Fig. 6 and the hard-core limit presented in Fig. 3(b) as both correspond to the same hysteresis protocol which give the same qualitative behaviour. Experimentally, the branches in Fig. 5 can be accessed through changing the direction of the drive $f$, as commonly experimentally realized to detect bi-stabilities in conventional polaritons [33–35]. The states with complex hysteresis protocols are more challenging to access as it requires to
break the translational invariance.

V. EXPERIMENTAL PERSPECTIVES AND CONCLUSIONS

We have shown that non-local interactions $V_{nn}$ have significant qualitative effects on the non-linear optical response of exciton-polaritons in moiré materials. In addition to the unique moiré-induced non-linearities arising of the strong on-site interactions [22, 25], the non-local interactions reveal new features including steady states with broken translational symmetry, multi-stabilities, and deviations from the on-site multi-photon resonance conditions. To study these features, we developed a self-consistent master equation based on a supercell containing three sites for excitons. These were treated independently at the mean-field level, allowing for the derivation of three coupled local master equations, which were solved self-consistently.

The predicted effects of the non-local interactions are readily measurable in experiment. They lead to a hysteretic dependence of the multi-photon lasing in the presence of non-local light-matter coupling. Another possibility is the study of excitonic states, the strong exciton interactions, and the interplay between multiple excitons in different contexts. For instance, an intriguing avenue is to understand the interplay between multiple excitonic states, wherein the form and position of the lobular pattern is determined by the direction of the drive (Fig. 8). Furthermore, a multi-valued hysteretic behaviour is found as a consequence of a spatial ordering of the excitons in the moiré sites. The form of these steady states could be experimentally detected by spatially resolving the positions of the excitons [36], but their existence is also apparent in measurements of the multi-stable hysteretic states of the cavity field [33, 35] (see Fig. 4).

Moiré systems are versatile platforms that allow for the control and manipulation of the excitonic properties over a wide range of parameters and our study encourages further studies to understand the behaviour of moiré excitons in different contexts. For instance, an intriguing avenue is to understand the interplay between multiple excitonic states, the strong exciton interactions, and the light-matter coupling. Another possibility is the study of the multi-photon lasing in the presence of non-local interactions [25]. In addition, the role of free carriers has been demonstrated to be a powerful tool with which to control optical non-linearities in van der Waals heterostructures [37–41]. The formation of moiré trions has been recently experimentally reported [9, 16] and the nature of the optical signatures of trion-polaritons stands as an interesting open question [42].

VI. ACKNOWLEDGMENTS

We thank Atac Imamoglu for the careful reading of the manuscript and valuable comments. This work was partially supported by EPSRC Grant Nos. EP/P009565/1, EP/P034616/1 and by a Simons Investigator Award. ACG acknowledges grant No. IN108620 from DGAPA (UNAM).

Appendix A: Dipolar interactions

We start discussing our approach for the non-local exciton-exciton interactions. Here, we assume that the non-local terms can be treated at the mean-field level, thus we have

$$\sum_{i \neq j} \frac{V_{ij}}{2} \hat{\mathbf{x}}_i \cdot \hat{\mathbf{x}}_j \rightarrow \sum_{i \neq j} \frac{V_{ij}}{2} \langle \hat{n}_i \rangle \langle \hat{n}_j \rangle + \sum_{i \neq j} V_{ij} \hat{n}_i \langle \hat{n}_j \rangle,$$

(A1)

that is, we take $\langle (\hat{n}_i - \langle n_i \rangle) (\hat{n}_j - \langle n_j \rangle) \rangle \approx 0$. Only the last term in (A1) will be relevant to the effective mean-field dynamics of the site $i$.

As illustrated in Fig. 7 we introduce three exciton sites that colour the moiré lattice. Thus, we define a larger supercell of three sites (labelled by $\alpha = R, G, B$) and denote the position of that supercell by the index $I$ such that the site index $i \rightarrow (I, \alpha)$. In our mean-field ansatz the occupations of sites of the same colour $\alpha$ within all supercells are equivalent $\langle \hat{n}_{(I, \alpha)} \rangle = n_{\alpha}$. Any given site interacts with sites of the same and different colours. In Fig. 7 we illustrate the non-local dipole-dipole interaction between a green site and (a) other green sites distanced by $r_{nm}^{gr} = |n \mathbf{a}_1 + m \mathbf{a}_2|$, from the original green site, (b) blue sites distanced by $r_{nm}^{rb} = |n \mathbf{a}_1 + m \mathbf{a}_2 + (\mathbf{a}_1 + \mathbf{a}_2)/3|$, where $n, m \in \mathbb{N}$. Here $\mathbf{a}_1 = a_M (\sqrt{3}, 0)$ and $\mathbf{a}_2 = a_M (3/2, \sqrt{3}/2)$ being $a_M$ the moiré lattice constant. [The red sites (not shown) are separated by $r_{nm}^{gr} = |n \mathbf{a}_1 + m \mathbf{a}_2 - (\mathbf{a}_1 + \mathbf{a}_2)/3|$.]

In detail, the interaction between a site $I$ with colour $\alpha$ and sites with the same colour $\alpha$ is given by

$$\sum_{J \neq I} V_{(I, \alpha), (J, \alpha)} \hat{n}_{(I, \alpha)} \langle \hat{n}_{(J, \alpha)} \rangle = \sum_{J \neq I} V_{(I, \alpha), (J, \alpha)} \hat{n}_{(I, \alpha)} \langle \hat{n}_{(J, \alpha)} \rangle$$

$$= n_{\alpha} \hat{n}_{I, \alpha} V_{nn} \sum_{n,m} \frac{a_M^3}{|n \mathbf{a}_1 + m \mathbf{a}_2|^3} \approx n_{\alpha} \langle \hat{n}_{(I, \alpha)} \rangle V_{nn} \times 2.12,$$

(A2)

where the sum in the second line is restricted to exclude $(n, m) = (0, 0)$. Here, $V_{nn} = d^2/a_M^3$. 

FIG. 7. (a) Non-local interaction between one green site (circled) and sites labelled with the same colour (b) Interaction between the green site (circled) and blue sites.
Similarly, the interaction between the site in supercell $I$ with colour $\alpha$ and all sites with a different colour $\beta \neq \alpha$ is

$$\sum_j V_{(I,\alpha),(j,\beta)} \hat{n}_{(I,\beta)} = \sum_j V_{(I,\alpha),(j,\beta)} \hat{n}_{(I,\beta)} n_{\beta}$$

$$\approx n_{\beta} \hat{n}_{(I,\beta)} V_{nn} m_{n} \left| a_m^I \right|^2$$

$$= n_{\beta} \hat{n}_{(I,\beta)} V_{nn} \times 4.455,$$

where the sign $\pm$ determines the colour of the sites. In Fig. 7 the sign $\pm$ determines the coupling of the green sites to the blue (+) red (−) sites respectively, both sums give the same factor of 4.455.

Then, for the site $\alpha$ within any supercell we can define a local Hamiltonian that includes the non-local exciton-exciton interactions at the mean-field level,

$$\left( V_q n_{\alpha} + V_{od} \sum_{\beta \neq \alpha} n_{\beta} \right) \hat{n}_{(I,\alpha)}$$

with $V_q = 2.124 \times V_{nn}$ and $V_{od} = 4.455 \times V_{nn}$ as denoted in the main text. There, the number operator is written $\hat{n}_{(I,\alpha)} = \hat{x}_{\alpha}^I \hat{\alpha}$, dropping the label $I$ of the supercell, since the mean-field self-consistency equation is the same for all supercells.

**Appendix B: Self-consistent scheme**

Our self-consistent scheme consists of the exact diagonalization of three Lindblad operators $L_\alpha$ for $\alpha = \text{R,G}$ and $\text{B}$ that are coupled through the cavity field $\psi_\alpha$ and the long-range interaction term of the dipole-dipole interactions. The self-consistent approach consists of iteratively obtaining the exciton coherences $\langle \hat{x}_\alpha \rangle = x_\alpha$ and the populations $n_\alpha$.

Numerically, the branches are obtained as follows.

1. For a given $\Delta \omega_X$ we start from a large $f$ and a random set of parameters $(x_\alpha^0, n_\alpha^0)$.

2. We evaluate $L_\alpha(x_\alpha^0, n_\alpha^0)$ and calculate $(x_\alpha^1, n_\alpha^1)$, which can lead in principle to different values than $(x_\alpha^0, n_\alpha^0)$.

3. We iterate $L_\alpha(x_\alpha^i, n_\alpha^i)$ using the parameters $(x_\alpha^{i-1}, n_\alpha^{i-1})$. We iterate up to $i = N = 1800$ or when $\max(\text{error}_R, \text{error}_G, \text{error}_B) < 10^{-8}$ where $\text{error}_\alpha = |n_{\alpha}^{i-1} - n_{\alpha}^i|$. The final state is denoted by $(x_\alpha(f, \Delta \omega_X), n_\alpha(f, \Delta \omega_X))$.

4. By keeping fixed $\Delta \omega_X$ we decrease $f$ by $\Delta f$. Depending on the branch we take $(x_\alpha^0, n_\alpha^0)$ as following:

- **Full hysteresis:**

  $x_\alpha^0(f - \Delta f, \Delta \omega) = x_\alpha(f, \Delta \omega_X),$

  $n_\alpha^0(f - \Delta f, \Delta \omega) = n_\alpha(f, \Delta \omega_X),$

  for $\alpha \in \{\text{R,G,B}\}$ That is, all of the results obtained for $(f, \Delta \omega_X)$ are employed. This protocol is used for Fig. 8(c) and the high-density hysteresis branch in Fig. 2.

- **Two excitons hysteresis:** We retain only two solutions, that is, we take for instance

  $x_\alpha^0(f - \Delta f, \Delta \omega) = x_\alpha(f, \Delta \omega_X),$

  $n_\alpha^0(f - \Delta f, \Delta \omega) = n_\alpha(f, \Delta \omega_X),$

  $x_\beta^0(f - \Delta f, \Delta \omega) = 0,$

  $n_\beta^0(f - \Delta f, \Delta \omega) = 0,$

  for $\alpha \in \{\text{R,G}\}$.

- **Single exciton hysteresis.** We retain only one solution, while the remaining needed parameters are set to zero. For instance, one of these branches corresponds to

  $x_\alpha^0(f - \Delta f, \Delta \omega) = x_\alpha^G(f, \Delta \omega_X),$

  $n_\alpha^0(f - \Delta f, \Delta \omega) = n_\alpha^G(f, \Delta \omega_X),$

  $x_\beta^0(f - \Delta f, \Delta \omega) = x_\beta^0(f - \Delta f, \Delta \omega) = 0,$

  $n_\beta^0(f - \Delta f, \Delta \omega) = n_\beta^0(f - \Delta f, \Delta \omega) = 0,$

  we use this procedure of Fig. 8(b) and Fig. 3(b).

- **Lower branch:** None of the solutions are kept, that is,

  $(x_\alpha^0(f - \Delta f, \Delta \omega), n_\alpha^0(f - \Delta f, \Delta \omega)) = (0, 0),$ for $\alpha \in \{\text{R,G,B}\}$. This is the scheme followed for the lower branch in Fig. 2 and Fig. 8(a).

5. We repeat step 2.

The convergence of our numerics is illustrated in Figs. 2, 3, 4 and Fig. 6 where error bars have been added and correspond to $|n_{(N)}(f, \Delta \omega_X) - n_{(N-1)}(f, \Delta \omega_X)|$. The barely visible error bars confirm that our results are fully converged.

**Appendix C: Hysteresis schemes**

The different hysteresis schemes permit multi-stabilities. As mentioned in the main text, the low- and high-density hysteresis branches in Fig. 5 are accompanied by two additional branches that can be accessed via the mechanisms discussed in the Appendix B. These solutions are illustrated in Fig. 8.

The steady states in Figs. 8 do not break translational symmetry, however, the stark difference between the phase-diagrams strongly depend on the initial seed of our numerics. Figure. 8(b) corresponds to the high-density hysteresis branch for a single coloured exciton.
FIG. 8. Exciton number per site for steady states with balanced colour population. (a) Low-density branch. (b) One exciton hysteresis protocol, (c) Two exciton hysteresis protocol and (d) full hysteresis scheme as explained above.

Figure. 8(c) corresponds to the hysteresis where two coloured excitons are recursively iterated. In addition, the deformation of the multi-photon resonances becomes more visible.

Finally, in Fig. 9 we set a much smaller value of the non-local interactions $V_{ad}/U_X$ to demonstrate that the local multi-photon resonances of Eq. 14 are recovered in this limit.

FIG. 9. Exciton number per site for steady states with balanced colour population. Here we illustrate the (a) low-density and (b) full hysteresis for $V_{ad}/U_X = 0.05$ and remaining same parameters as in Fig. 8.

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