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

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(Dated: October 26, 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 spontaneously 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 [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 [2], Mott insulators [3], superconductivity [4, 5] and more [6–11]. In semiconductor bilayers, moiré materials have unveiled a new class of excitations [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 [17, 18], the superfluid-Mott transition [19], excitonic insulators [20], and supersolidity [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 [22], novel forms of quantum emitters [23–25], and promises a new generation of polaritons with tuneable features [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 [12, 28] and strong exciton-exciton interactions [22]. Recent experimental [22] and theoretical studies [25] have demonstrated that moiré polaritons feature optical properties with large non-linearities very different from conventional polaritons in semiconductors. 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 [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. The role of non-local interactions on out-of-equilibrium polaritons is further motivated by theoretical predictions and breakthrough experiments where non-local interactions are a key element to stabilizing states with spontaneously broken translational symmetry which can lead to complex many-body phases such as supersolids [29–32], and which have been already been observed in dipolar quantum gases [33–35]. The dipole-dipole interaction between excitons beyond the local interaction has very recently suggested the existence of supersolid phases of moiré excitons [21], and instigates the study of the interplay between these phases and polariton physics.

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 hardcore 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 have 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

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

(1)

here \( \hat{x}_i^\dagger \) creates an exciton with energy \( \omega_X \) in the site \( i \), with \( N_x \) 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

\[ \hat{H}_l = \sum_{\mathbf{k}} \omega_c(\mathbf{k}) \hat{a}^\dagger_{\mathbf{k}} \hat{a}_\mathbf{k}. \]

(2)

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

\[ \hat{H}_{1-m} = \sum_{\mathbf{k}} \Omega \left( \hat{a}^\dagger_{\mathbf{k}} \hat{x}_\mathbf{k} + \hat{x}^\dagger_{\mathbf{k}} \hat{a}_\mathbf{k} \right), \]

(3)

where \( \hat{x}^\dagger_{\mathbf{k}} \) creates an exciton with in-plane momentum \( \mathbf{k} \).

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.) We, however, consider the regime of strong light-matter coupling such that the Rabi coupling is much larger than the cavity losses \( \gamma_c \), and light couples efficiently to the excitons.

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

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

(4)

where \( \hat{a} \) now refers to the \( \mathbf{k} = 0 \) 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
\[ \hat{H}_{\text{drive}} = (F \hat{a}^\dagger e^{-i \omega_{\text{p}} t} + F^* \hat{a} e^{i \omega_{\text{p}} t}), \]
where \( F \) and \( \omega_{\text{p}} \) 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 \( \mathbf{k} = 0 \) cavity mode. We describe the system in the rotating frame of this light field, using \( \hat{a} = \hat{a} e^{i \omega_{\text{p}} t} \) and \( \hat{x}_k = \hat{x}_k e^{i \omega_{\text{p}} t} \), for simplicity we drop the \( \sim \)’s in the following. The rotating frame introduces the pump energy detuning from the exciton and cavity detuning defined as \( \Delta \omega_X = \omega_p - \omega_X \) and \( \Delta \omega_c = \omega_p - \omega_c \), respectively.

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 \cite{36, 37}, such effects can be accounted to a first approximation through an anharmonic light-matter coupling term \cite{38–40}. This term, however, only quantitatively modifies the optical properties for small exciton numbers \cite{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 \cite{41}
\[ \frac{d\hat{\rho}}{dt} = -i[\hat{H}, \hat{\rho}] + \mathcal{D}[\hat{\rho}] = \mathcal{L}[\hat{\rho}], \]
where the total Hamiltonian is given by \( \hat{H} = \hat{H}_X + \hat{H}_I + \hat{H}_{\text{loc}} + \hat{H}_{\text{drive}} \). The dissipative character of the system is accounted for by the operator
\[ \mathcal{D}[\hat{\rho}] = \frac{\gamma_c}{2} \left[ 2 \hat{a} \hat{a}^\dagger \hat{\rho} - \{\hat{\rho}, \hat{a}^\dagger \hat{a}\} \right] + \sum_x \frac{\gamma_x}{2} \left[ 2 \hat{x}_x \hat{x}_x^\dagger \hat{\rho} - \{\hat{\rho}, \hat{x}_x^\dagger \hat{x}_x\} \right], \]
where \( \gamma_x \) 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 only consider the three 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. \cite{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. \( \ref{eq:hamiltonian} \). 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 \( \hat{x}_i \rightarrow \hat{x}_\alpha \) where \( \alpha \in \{\text{R,G,B}\} \) labels the three distinct sites within the supercell. The Hamiltonian for each colour is then
\[ \hat{H}_{X,\alpha} = \left( \omega_X + V_{\text{d}a} n_\alpha + V_{\text{od}} \sum_{\beta \neq \alpha} n_\beta \right) \hat{\hat{x}}^\dagger_\alpha \hat{x}_\alpha + \frac{U_X}{2} \hat{x}_\alpha^\dagger \hat{x}_\alpha^\dagger \hat{x}_\alpha \hat{x}_\alpha + \frac{U_{X\text{d}}}{4} \hat{\hat{x}}^\dagger_\alpha \hat{\hat{x}}^\dagger_\alpha \hat{x}_\alpha \hat{x}_\alpha, \]
this local Hamiltonian treats the local terms in Eq. \( \ref{eq:hamiltonian} \) 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, \( V_{\text{d}a} = 2.124 \times V_{nn} \) and \( V_{\text{od}} = 4.455 \times V_{nn} \) 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 and Ref. \cite{42}. Here, \( V_{nn} = d^2/a_M^3 \) is the dipole-dipole interaction between excitons in nearest neighbour sites, where \( a_M \) is the moiré period of the superlattice and \( d \) is the dipole moment of the hybrid exciton due to its interlayer charge separation.

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
\[ \hat{H}_{1-m,\alpha} = \frac{\Omega}{\sqrt{N_s}} \left( \hat{\hat{a}}^\dagger \hat{x}_\alpha + \hat{\hat{a}} \hat{x}_\alpha^\dagger \right). \]
To make further progress, we take a semi-classical approach for the cavity photons, where we replace the cavity field by its expectation value \( \langle \hat{a} \rangle = \sqrt{N_s} \psi_a \). In this case, the steady-state solution for the photon amplitude \( \psi_a \) is given by
\[ \psi_a = \frac{1}{\Delta_{\text{c}}} \left( f + \frac{\Omega}{3} \sum_{\alpha = \text{R,G,B}} \langle \hat{x}_\alpha \rangle \right), \]
where \( f = F/\sqrt{N_s} \). The last term inside of the brackets accounts for the possible different expectation value of sites with different colour. Here, \( \Delta_{\text{c}} = \Delta \omega_{\text{c}} + i \gamma_{\text{c}}/2 \).

These approximations permit us to define three Hamiltonians for differently colours \( \alpha \),
\[ \hat{H}_{\text{loc},\alpha} = \hat{H}_{X,\alpha} + \hat{H}_{1-m,\alpha}, \]
with \( \alpha, \beta \in \{\text{R,G,B}\} \), 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 \( V_{\text{d}a} \) and \( V_{\text{od}} \), in Eq. \( \ref{eq:hamiltonian} \) treated at the mean-field level. This mean-field approximation can be understood intuitively: it introduces a self-consistent on-site energy that can vary for the three different colours and thus, can energetically favour the breaking of the translational symmetry. That is, the imbalanced-population steady-state solutions are a consequence of an emergent
colour-dependent on-site energy arising from the non-local interactions. We emphasize that since our starting Hamiltonian has the full translational symmetry of the triangular lattice, the emergence of collective phases with a reduced translational symmetry through interactions is an example of spontaneously broken symmetry [43]. Thus there is a residual (discrete) degeneracy of the ground state associated with the different ways in which these broken symmetry states can be placed on the triangular lattice. Which of the broken-symmetry states appears in the numerics is, as usual, determined by the initial seed. On the other hand, 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. That is, we assume 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

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

\[= -\frac{i}{2} [\hat{H}_{\text{loc},\alpha}, \hat{\rho}_\alpha] + \frac{\gamma_\alpha}{2} \left( 2\hat{x}_\alpha \hat{\rho}_\alpha \hat{x}_\alpha^\dagger - \{\hat{x}_\alpha^\dagger \hat{x}_\alpha, \hat{\rho}_\alpha\} \right),\]

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 $\psi_\alpha$ and the exciton number $\langle x_\alpha^\dagger x_\beta \rangle$ 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 [22], the interaction between first neighbours is estimated $V_{nn} \sim 0.1 - 1$ meV [21], 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 interactions over a wide range of parameters. We expect our approximation to be valid when the non-local interaction remains smaller compared to the on-site interaction $V_a/U_X \ll 1$ and $V_{aa}/U_X \ll 1$. 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 occupa-


tion, 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.

![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)

The behaviour just described appears qualitatively similar to that found in Ref. [25], where it was shown 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 an apparent bistability.

Intriguingly, as we now show, in this regime we find that the presence of non-local interactions can lead to additional steady-state solutions, turning the bistability into a multi-stability with four steady-states: the two solutions in Eq. 2 and two additional solutions with spatial ordering of the excitons, which we will now discuss. These solutions are illustrated in Fig. 3 for the same values of the parameters as in Fig. 2.

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.

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 $n_p = |\psi|^2$ 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.

FIG. 3. Exciton number per site. (a) Steady-state solutions with $n_B = n_C > n_R$, 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. Color code follows the lattice colouring. Parameters are the same as for Fig. 2.

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 population 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, allowing for fully population-imbalanced metastable states with $n_R \neq n_B, n_B \neq n_G$, and $n_R \neq n_G$, but our calculations show that these do not appear over the wide range of parameters that we have explored. We speculate that the absence of these solutions arises from the fact that each site can transit either to a low- or high-density phase, this binary characterisation of the population of the sites yields to only four kinds of different solutions shown in Fig. 2 and Fig. 3.

The size of the pink area is determined by the ratio between the light-matter coupling and the strength of the non-local interactions. To illustrate this point we show in Fig. 4 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 disappear 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 spontaneously broken translational symmetry, that is, with unbalanced population in terms of the exciton colour. Numerically, we
access states with broken translational symmetry by ini-
itializing our self-consisting approach with seeds that ex-
plicitly break this symmetry, see further details in the
Appendix B. We stress that although the ability to access
states with broken translational symmetry depends on
the initial seed, the existence of such steady-states hinges
on the non-local interactions and as these states are ab-
sent when $\Omega/V_{nn} \gg 1$. This dependence on the initial
seed is a feature commonly shared for any broken sym-
metry phenomenon, where the initial seed is used only
to select which of the various broken symmetry states is
realized. For example, in typical self-consistent schemes
for equilibrium Bose-Hubbard-like models, the detection
of states with broken translational symmetry requires an
initial ansatz that explicitly breaks this symmetry; then,
the self-consistent approach can drive the solution into
equilibrium states that may preserve the broken transla-
tional symmetry.

Our ansatz is based on the uses of a supercell contain-
ing three kinds of moiré sites, thus, the solutions follow
the restriction imposed by that ansatz. Solutions with
different spatial structures, not allowed by our ansatz,
could emerge through the use of other supercells. We
have also studied another simple two-site supercell which
allows for striped density order. We find that striped
solutions can also be appeared, but that these are less
stable than the structures that we present here. In par-
ticular, the striped phases do not appear for the set of
parameters discussed in Figs. 1-3. We cannot rule out
that other more complex spatial structures, which are
possible only in larger supercells, could be more stable
than those we present for the 3-site cell. Also, we expect
that disorder could play a significant role in determin-
ing the nature of the stable states. For clarity of our
presentation, we leave the comprehensive study of more
complex broken symmetry phases and disorder to future
investigations.

IV. MULTI-PHOTON RESONANCES AND
NON-LOCAL INTERACTIONS

Now, we turn our attention to the study of the inter-
play 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

$$N\omega_p = N\omega_X + \frac{U_X}{2}N(N-1), \quad (13)$$

which leads to the condition

$$\frac{2\Delta\omega_X}{U_X} = (N-1). \quad (14)$$

Physically this can be understood in terms of an ener-
geic condition dictating that $N$ exciton resonances are
promoted whenever the energy of $N$ non-interacting pho-
tons matches the energy of $N$ interacting excitons [25]. 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

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

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

$$N\omega_p = N\left(\omega_X + V_d n_\alpha + V_{od} \sum_{\beta \neq \alpha} n_\beta\right) + \frac{U_X}{2}N(N-1). \quad (15)$$

In this case, the multi-photon resonance (14) depends on
$n_\beta$ and is not longer necessarily an integer. One should
note that in contrast to the Bose-Hubbard model in equi-
librium where the occupation number per site of the in-
sulating phase is pinned to integer values, for driven-
dissipative systems the discrete lobular pattern deter-
mined by the multi-photon resonance condition is a rem-
nant of the discreteness of the equilibrium Hubbard en-
ergy spectrum. However, the occupation number is no
longer strictly an integer. That is, while the modulation
of the phase diagram for $V_{nn} = 0$ follows very closely the
discrete equation in Eq. 13, the exact value of the exciton
number slightly above the low-to-high density transition is
not 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 corre-
sponds to $\Omega/V_{nn} = 1.42$, finally, we consider $V_{od}/U_X = \frac{54x}{70}$

![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 $V_{od}/U_X \approx 0.1$, $\Omega/U_X = 0.65$, $\Delta\omega_c/U_X = -1$ and $\gamma_c/U_X = 0.2$.](image-url)
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 \((n_R = n_B = n_{Q_2})\) 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. 10 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 [40, 41, 44]. In general the states with complex hysteresis protocols are more challenging to access, as which of the states of differently broken translational symmetry will appear depends on how the translational symmetry breaking is seeded — by preparation or through underlying disorder.

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 hys-
teretic dependence of the multi-photon resonance condition, wherein the form and position of the lobular pattern is determined by the direction of the drive (Fig. 9). 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 [45], but their existence is also apparent in measurements of the multi-stable hysteretic states of the cavity field [40, 44] (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, for instance, by twisting the relative angle between the layers or by inserting an insulating layer between them to control the moiré superlattice properties and the features and degree of hybridisation of the excitons [46]. 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 non-local terms can be treated at the mean-field level, such that we take

\[ \sum_{i} V_{ij} \hat{n}_i \hat{n}_j \approx \frac{1}{a_M^3} \left( n_a + n_b + n_c \right)^3 \]

that is, we take \( \langle \hat{n}_i - \langle n_i \rangle \rangle \approx 0 \). 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 \( r_{n,m}^{gg} = |n\mathbf{a}_1 + m\mathbf{a}_2| \), from the original green site, (b) blue sites distanced by \( r_{n,m}^{gb} = |n\mathbf{a}_1 + m\mathbf{a}_2 + (\mathbf{a}_1 + \mathbf{a}_2)/3 \), where \( n, m \in \mathbb{Z} \). Here \( a_1 = a_M(\sqrt{3}, 0) \) and \( a_2 = a_M(3/2, \sqrt{3}/2) \) being \( a_M \) the moiré lattice constant. The red sites (not shown) are separated by \( r_{n,m}^{gr} = |n\mathbf{a}_1 + m\mathbf{a}_2 - (\mathbf{a}_1 + \mathbf{a}_2)/3 | \).

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

\[ \sum_{J \neq I} V_{I,\alpha}(J, \alpha) \hat{n}_{I,\alpha}(J, \alpha) \hat{n}_{I,\alpha} = \frac{a_M^3}{|n_1 + n_2|^3} \sum_{n,m} V_{nn} n_\alpha \hat{n}_{I,\alpha} \hat{n}_{I,\alpha} \approx n_\alpha \hat{n}_{I,\alpha} V_{nn} \times 2.12, \]

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

Similarly, the interaction between the site in supercell \( I \) with colour \( \alpha \) and all sites with a different colour \( \beta \neq \alpha \) is

\[ \sum_{J} V_{I,\alpha}(J, \beta) \hat{n}_{I,\alpha}(J, \beta) \hat{n}_{I,\alpha} \approx n_\alpha \hat{n}_{I,\alpha} V_{nn} \sum_{n,m} a_M^3 |n_1 + n_2 + \alpha_1 + \alpha_2/3|^3 \approx n_\beta \hat{n}_{I,\alpha} V_{nn} \times 4.455, \]

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; these sums are evaluated numerically and agree with the known results from Ref. [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

\[ \sum_{i} \frac{V_{ij}}{2} \hat{n}_i \hat{\phi}_j + \sum_{i} \frac{V_{ij}}{2} \langle \hat{\phi}_i \rangle \langle \hat{\phi}_j \rangle + \sum_{i \neq j} V_{ij} \hat{n}_i \hat{n}_j, \]

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

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

with $V_d = 2.124 \times V_{nn}$ and $V_{od} = 4.455 \times V_{nn}$ as denoted in the main text. There, the number operator is written $\hat{n}_{(I,\alpha)} = \hat{x}_{\alpha}^\dagger \hat{x}_\alpha$, 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

We now provide the details of our self-consistent approach which is based on an iterative exact diagonalization of the three Lindblad operators $\mathcal{L}_\alpha$ for $\alpha = R,G$ and $B$ 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 $n_\alpha$.

The iterative scheme is obtained as follows and illustrated in Fig. 8 for a particular hysteresis branch.

1. For a given $\Delta\omega_X$ we start from a large $f_0$ where the steady-state is single valued. Thus, we start from a random set of parameters $(x_0^0(f_0), n_0^0(f_0))$. Here, the super-index denotes the step of the iteration which we now discuss.

2. We calculate $\mathcal{L}_\alpha(x_0^0(f_0), n_0^0(f_0))$ and calculate the expectation values of the operators $\hat{x}_\alpha$ and $\hat{n}_\alpha$ which define the seed for the next iteration $(x_{1,\alpha}^*(f_0), n_{1,\alpha}^*(f_0))$, which interpolates $x_\alpha^0(f_0) = \langle \hat{x}_\alpha \rangle = \langle \hat{x}_\alpha \rangle - \eta(\langle \hat{x}_\alpha \rangle - x_0^0(f_0))$, where $\eta$ is adjusted to speed numerical convergence. Note that the sub-index has remained unchanged.

3. We then iterate $\mathcal{L}_\alpha(x_{i,\alpha}^*(f_0), n_{i,\alpha}^*(f_0))$ using the parameters $(x_{i-1,\alpha}^0(f_0), n_{i-1,\alpha}^0(f_0))$. We iterate up to $i = N_{\text{max}} = 1800$ or when max(error$_R$,error$_G$,error$_B$) $< 10^{-8}$ where error$_\alpha = |n_{i-1,\alpha} - n_{i,\alpha}|$. The final state is denoted by $(x_{N_{\text{iter}},\alpha}^0(f_0), n_{N_{\text{iter}},\alpha}^0(f_0))$, where $N_{\text{iter}}$ is either $N_{\text{max}}$ or the number of iteration steps required to converge. Steps 2-3 are used for all of our numerics.

4. After convergence is achieved for $f_0$, we then decrease $f_0$ by an amount of $\Delta f > 0$, we define $f_1 = f_0 - \Delta f$. The initial seed $(x_0^0(f_1), n_0^0(f_1))$ is no longer random and is taken as detailed below:

- Full hysteresis:
  $$
x_0^0(f_1) = x_{N_{\text{iter}},\alpha}^0(f_0),
  
n_0^0(f_1) = n_{N_{\text{iter}},\alpha}^0(f_0),
$$

for $\alpha \in \{R,G,B\}$ That is, all of the results obtained for $f_0$ are preserved. This protocol is used for Fig. 9(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_0^0(f_1) = x_{N_{\text{iter}},\alpha}^0(f_0),
  
n_0^0(f_1) = n_{N_{\text{iter}},\alpha}^0(f_0),
  
x_0^0(f_1) = 0,
  
n_0^0(f_1) = 0,
$$

for $\alpha \in \{R, G\}$. We use this scheme in Fig. 3(a) and Fig. 9(c).

- 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_0^0(f_1) = x_{N_{\text{iter}},\alpha}^0(f_0),
  
n_0^0(f_1) = n_{N_{\text{iter}},\alpha}^0(f_0),
  
x_0^0(f_1) = 0,
  
n_0^0(f_1) = 0,
$$

we use this procedure of Fig. 9(b) and Fig. 3(b). The procedure for this branch is illustrated in Fig. 8.

- Lower branch: None of the solutions are kept, that is,
  $$
  (x_0^0(f_1), n_0^0(f_1)) = (0, 0),
$$

for $\alpha \in \{R,G,B\}$. This is the scheme followed for the lower branch in Fig. 2 and Fig. 9(a).

5. We repeat step 2. Again, the arbitrary initial seed is only used to follow each hysteresis branch.

The convergence of our numerics is illustrated in Figs. 2, 3, 4 and Fig. 6 where error bars have been added and correspond to $|n_{N_{\text{iter}},\alpha}(f) - n_{N_{\text{iter}}-1}(f)|$. The barely visible error bars confirm that our results are fully converged. For Sec. III, the Hilbert space of the excitons is naturally restricted to having at most one exciton per site. For finite on-site interactions we introduce a cut-off and restrict to ten excitons per site. The validity of this truncation depends on the strength of the drive and the exciton detuning that determine the exciton number. For the spanned parameters we restrict to exciton occupations much smaller than our cut-off and have indeed verified that our results do not change for larger sizes. Restricting to relatively small occupation number is also motivated by the experimental limitations to create arbitrary numbers of excitons per site through effects such as saturation, population of higher bands, or even experimental damage of the samples due to the high intensity of the laser.
The different hysteresis schemes permit multistabilities. 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. 9.

The steady states in Figs. 9 do not break translational symmetry, however, the stark difference between the phase-diagrams strongly depend on the initial seed of our numerics. Figure. 9(b) corresponds to the high-density hysteresis branch for a single coloured exciton.

Figure. 9(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. 10 we set a much smaller value of the non-local interactions $V_{od}/U_X$ to demonstrate that the local multi-photon resonances of Eq. 14 are recovered in this limit.

Appendix C: Hysteresis schemes

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FIG. 9. 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.

FIG. 10. Exciton number per site for steady states with balanced colour population. Here we illustrate the (a) low-density and (b) full hysteresis for $V_{od}/U_X = 0.05$ and remaining same parameters as in Fig. 9.

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