Cold Atoms and Molecules in Self-Assembled Dipolar Lattices

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We study the realization of lattice models, where cold atoms and molecules move as extra particles in a dipolar crystal of trapped polar molecules. The crystal is a self-assembled floating mesoscopic lattice structure with quantum dynamics given by phonons. We show that within an experimentally accessible parameter regime extended Hubbard models with tunable long-range phonon-mediated interactions describe the effective dynamics of dressed particles.

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Trapped atomic and molecular quantum gases allow the realization of quantum lattice models of strongly interacting bosonic and fermionic particles. For example, the dynamics of atoms in optical lattices is well described by a Hubbard model, where the tunability of the Hubbard parameters via external fields combined with atomic physics techniques of preparation and measurement provides a *quantum simulator* of strongly correlated condensed matter models [1]. In this Letter we propose and study an alternative scenario of realizing lattice models, where a dipolar crystal of trapped polar molecules provides a self-assembled floating lattice structure for extra particles, which are atoms or molecules of a second species (Fig. 1). By confining polar molecules to an effective 2D [Fig. 1(a)] or 1D [Figs. 1(b) and 1(c)] geometry by strong transverse trapping, dipolar crystals can form as a result of the balance of strong repulsion between the dipoles aligned by an external electric field, and an in-plane trapping potential [2,3]. Particles moving in this lattice under conditions of elastic scattering see a periodic potential, and thus form a lattice gas.

The distinguishing features of this realization of lattice models are (i) dipolar molecular crystals constitute an array of microtraps with its own quantum dynamics represented by phonons (lattice vibrations), while the lattice spacings are tunable with external control fields, ranging from a μm down to the hundred nm regime, i.e., potentially smaller than for optical lattices, and (ii) the motion of the extra particles is governed by an interplay of Hubbard (correlation) dynamics in the lattice and coupling to phonons. The tunability of the lattice allows one to access a wide range of Hubbard parameters and phonon couplings. Compared with optical lattices, for example, a small scale lattice yields significantly enhanced hopping amplitudes, which set the relevant energy scale for our Hubbard model (e.g., for exchange interactions), and thus also the temperature requirements for realizing strongly correlated quantum phases. Hopping amplitudes can be modified by coupling to phonons, while phonon-mediated interactions realize extended Hubbard models with strong off-site interactions, which are difficult to achieve in optical lattices. While the setups we propose are reminiscent of (and may be relevant to) solid state systems with strong phonon couplings, as, e.g., polaronic and/or superconducting materials, they realize the unusual parameter regime where the mass of the crystal's and of extra particles can be comparable.

A homogeneous lattice of polar molecules underlying the configurations of Fig. 1 relies on the strong repulsive dipole-dipole interactions $V_c(\mathbf{R}) = d_c^2/R^3$ with R the distance between the molecules, and d_c the dipole moment induced by a transverse electric field $E_{\rm dc}$. A requirement for the existence of a crystal is that the ratio of the potential energy to the kinetic energy of small oscillations around the equilibrium position, $r_d = d_c^2 m_c/\hbar^2 a$, with a the lattice spacing and m_c the mass, is larger than a critical value r_c, where $r_c = 18 \pm 4$ and $r_c \sim 1$ for bosons at zero temperature in 2D and 1D, respectively [2]. Thus a dipolar crystal will form for $a < a_{\rm max} \equiv d_c^2 m_c/\hbar^2 r_c$. In addition, we have $a_{\rm min} < a$ with $a_{\rm min} = (12 d_c^2/m_c \omega_\perp^2)^{1/5}$, which reflects the

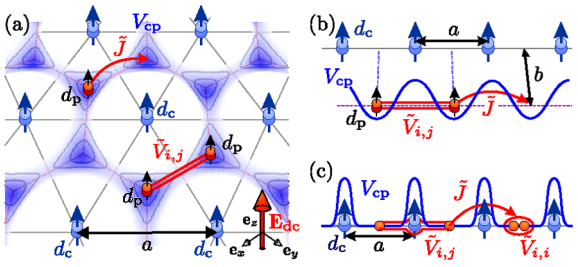

FIG. 1 (color online). A dipolar crystal of polar molecules in 2D (a) and 1D (b),(c) provides a periodic lattice $V_{\rm cp}$ for extra atoms or molecules giving rise to a lattice model with hopping $\tilde{J}$ and long-range interactions $\tilde{V}_{i,j}$ (see text). (a) In 2D a triangular lattice is formed by polar molecules with dipole moment d_c perpendicular to the plane. A second molecular species with dipole moment $d_p \ll d_c$ moves in the honeycomb lattice $V_{\rm cp}$ (darker shading corresponds to deeper potentials). (b) A 1D dipolar crystal with lattice spacing a provides a periodic potential for a second molecular species moving in a parallel tube at distance b (configuration 1). (c) 1D setup with atoms scattering from the dipolar lattice (configuration 2).

requirement of strong transverse trapping with a harmonic oscillator frequency $\omega_\perp$ to prevent collapse due to attractive forces between aligned dipoles [2]. For RbCs (SrO) molecules with permanent dipole moment $d_c = 1.27$ D ($d_c = 8.9$ D) confined by an optical lattice with $\omega_\perp/2\pi \sim 150$ kHz, $a_{\min} \sim 100$ nm (200 nm), while $a_{\max}$ can be several μm. Excitations of the crystal are acoustic phonons with Hamiltonian $H_c = \sum_q \hbar\omega_q a_q^\dagger a_q$, where a_q destroys a phonon of quasimomentum $\mathbf{q}$ in the mode λ. In 1D, $\hbar\omega_q = (2/\pi^2)[12 r_d f_q]^{1/2} E_{R,c}$ with Debye frequency $\hbar\omega_D \equiv \hbar\omega_{\pi/a} \sim 1.4\sqrt{r_d} E_{R,c}$, $f_q = \sum_{j>0} 4\sin(qaj/2)^2/j^5$, and lattice recoil frequency $E_{R,c} \equiv \hbar^2\pi^2/2m_c a^2$ (typically a few to tens of kHz). In 2D, $\hbar\omega_D \sim 1.6\sqrt{r_d} E_{R,c}$. The classical melting of the crystal into a normal phase occurs at $k_B T_C \simeq 0.2 r_d E_{R,c}$ ($0.018 r_d E_{R,c}$) in 1D (2D) [2,4].

An extra particle confined to the 2D crystal plane [Fig. 1(a)] or a 1D tube [Figs. 1(b) and 1(c)] will scatter from the periodic lattice potential $\sum_j V_{\rm cp}(\mathbf{R}_j - \mathbf{r})$ with $\mathbf{r}$ and $\mathbf{R}_j = \mathbf{R}_j^0 + \mathbf{u}_j$ the coordinates of the particle and crystal molecule j, respectively, with $\mathbf{R}_j^0$ the equilibrium positions and $\mathbf{u}_j$ small displacements. For particles being molecules, this potential is given by the repulsive dipole-dipole interaction $V_{\rm cp}(\mathbf{R}_j - \mathbf{r}) = d_p d_c/|\mathbf{R}_j - \mathbf{r}|^3$ with $d_p \ll d_c$ the induced dipole moment, and for atoms we assume that the interaction is modeled by a short range pseudopotential proportional to an elastic scattering length $a_{\rm cp}$. In addition, extra molecules and atoms will interact according to dipolar, or short range interactions, respectively.

We consider a situation where the extra particles in the lattice are described by a single band Hubbard Hamiltonian coupled to the acoustic phonons of the lattice [5]

$$H = -J\sum_{\langle i,j\rangle} c_i^\dagger c_j + \tfrac{1}{2}\sum_{i,j} V_{ij} c_i^\dagger c_j^\dagger c_j c_i + \sum_{q,j} M_q e^{i\mathbf{q}\cdot\mathbf{R}_j^0} c_j^\dagger c_j (a_q + a_{-q}^\dagger) + H_c. \quad (1)$$

The first and second terms describe the nearest-neighbor hopping of the extra particles with hopping amplitudes J, and interactions V, computed for each microscopic model by band-structure calculations for $\mathbf{u}_j = 0$, respectively (see below). We denote by c_i ($c_i^\dagger$) destruction (creation) operators of the particles. The third term is the phonon coupling obtained in lowest order in the displacement $\mathbf{u}_j = i\sum_q (\hbar/2m_c N\omega_q)^{1/2}\xi_q (a_q + a_{-q}^\dagger) e^{i\mathbf{q}\cdot\mathbf{R}_j^0}$ with $M_q = \bar{V}_\mathbf{q}\mathbf{q}\cdot\xi_q(\hbar/2Nm_c\omega_q)^{1/2}\beta_\mathbf{q}$, where ξ_q and N are the phonon polarization and the number of lattice molecules, respectively, $\bar{V}_\mathbf{q}$ is the Fourier transform of the particle-crystal interaction $V_{\rm cp}$, and $\beta_\mathbf{q} = \int d\mathbf{r}|w_0(\mathbf{r})|^2 e^{i\mathbf{q}\mathbf{r}}$, with $w_0(\mathbf{r})$ the Wannier function of the lowest Bloch band [5]. The validity of the single-band Hubbard model requires J, $V < \Delta$, and temperatures $k_B T < \Delta$ with Δ the separation to the first excited Bloch band. We note that the Hubbard parameters are of the order of magnitude of the recoil energy, J, $V \sim E_{R,c}$, and thus (much) smaller than the Debye frequency $\hbar\omega_D \sim E_{R,c}\sqrt{r_d}$, for $r_d \gg 1$ [6].

Below we will present detailed results for the examples of Figs. 1(b) and 1(c). The separation of time scales $J, V \ll \hbar\omega_D$, combined with the fact that the coupling to phonons is dominated by high frequencies $\hbar\omega > J$, V (see the discussion of M_q below) is reminiscent of polarons as particles dressed by (optical) phonons, where the dynamics is given by coherent and incoherent hopping on a lattice [5,7]. This physical picture is brought out in a master equation treatment within a strong coupling perturbation theory. The starting point is a Lang-Firsov transformation of the Hamiltonian $H \to \mathcal{S}H\mathcal{S}^\dagger$ with a density-dependent displacement $\mathcal{S} = \exp[-\sum_{q,j}(M_q/\hbar\omega_q) e^{i\mathbf{q}\mathbf{R}_j^0} c_j^\dagger c_j (a_q - a_{-q}^\dagger)]$. This eliminates the phonon coupling in the second line of Eq. (1) in favor of a transformed kinetic energy term $-J\sum_{\langle i,j\rangle} c_i^\dagger c_j X_i^\dagger X_j$, where the displacement operators $X_j = \exp[\sum_q M_q e^{i\mathbf{q}\mathbf{R}_j^0}(a_q - a_{-q}^\dagger)/\hbar\omega_q]$ can be interpreted as a lattice recoil of the dressed particles in a hopping process. In addition, the bare interactions are renormalized according to $\tilde{V}_{ij} = V_{ij} + V_{ij}^{(1)}$ with $V_{ij}^{(1)} = -2\sum_q \cos(\mathbf{q}(\mathbf{R}_i^0 - \mathbf{R}_j^0)) M_q^2/\hbar\omega_q$, that is, the phonon couplings induce and modify off-site interactions. The on-site interaction is given by $\tilde{V}_{j,j} = V_{j,j} - 2E_p$ with $E_p = \sum_q M_q^2/\hbar\omega_q$ the *polaron* self-energy or *polaron shift*. For $J = 0$ the new Hamiltonian is diagonal and describes interacting polarons and independent phonons. The latter are vibrations of the lattice molecules around new equilibrium positions with unchanged frequencies. A stable crystal requires the variance of the displacements Δu around these new equilibrium positions to be small compared to a [2,8].

A Born-Markov approximation with the phonons a finite temperature heat bath with J, $V \ll \hbar\omega_D$ (see above), and the transformed kinetic energy as perturbation [9], provides the master equation for the reduced density operator of the dressed particles ρ_t in Lindblad form [10]

$$\dot{\rho}_t = \frac{i}{\hbar}[\rho_t, \tilde{H}] + \sum_{j,l,\delta,\delta'} \frac{\Gamma_{j,l}^{\delta,\delta'}}{2\hbar}([b_{j\delta}, \rho_t b_{l\delta'}] + [b_{l\delta'}, \rho_t b_{j\delta}]), \quad (2)$$

with $b_{j\delta} = c_{j+\delta}^\dagger c_j$. The effective system Hamiltonian

$$\tilde{H} = -\tilde{J}\sum_{\langle i,j\rangle} c_i^\dagger c_j + \tfrac{1}{2}\sum_{i,j} \tilde{V}_{ij} c_i^\dagger c_j^\dagger c_j c_i, \quad (3)$$

is of the extended Hubbard type, valid for $\tilde{J}$, $\tilde{V}_{ij}$, $E_p < \Delta$. For $E_p > \Delta$, Eq. (3) should be derived via a multiband approach. *Coherent* hopping of the dressed particles is described by $\tilde{J} = J\langle\langle X_i^\dagger X_j\rangle\rangle \equiv J\exp(-S_T)$, where $S_T = \sum_q (M_q/\hbar\omega_q)^2[1 - \cos(\mathbf{q}\mathbf{a})](2n_q(T) + 1)$ characterizes the strength of the particle-phonon interactions, and $n_q(T)$ is the thermal occupation at temperature T [5,11].

The dissipative term in Lindblad form in Eq. (2) corresponds to thermally activated *incoherent* hopping with

rates $\Gamma_{j,l}^{\delta,\delta'}$. In 1D, the latter are small compared to $\tilde{J}$ for $S_T \ll 1$ and $S_T \gg 1$, provided $J\bar{V}_{q=0}^2 k_B T/[(\hbar\omega_D)^4 \times \sqrt{r_d}] \ll 1$ and $k_B T/E_p \ll 1$, respectively [7,12], and, in particular, they are negligible for the energies of interest $k_B T \ll \min(\Delta, E_p, k_B T_C)$.

In the parameter regime of interest the dynamics of the dressed particles is described by the extended Hubbard Hamiltonian $\tilde{H}$. In the following, we calculate the effective Hubbard parameters from the microscopic model for the 1D configurations described in Figs. 1(b) and 1(c) [13].

In *configuration 1* [Fig. 1(b)] molecules of a second species are trapped in a tube at a distance b from the crystal tube under 1D trapping conditions. For crystal molecules fixed at the equilibrium positions with lattice spacing a, the extra particles feel a periodic potential $V_{\rm cp}(x) = d_c d_p \sum_j [b^2 + (x - ja)^2]^{-3/2}$, which determines the band structure. The lattice depth $V_0 \equiv V_{\rm cp}(a/2) - V_{\rm cp}(0) \sim r_d (d_p m_p / d_c m_c) e^{-3b/a} E_{R,p}/(b/a)^3$ is shown in Fig. 2(a) as a function of b/a, where the thick solid lines indicate the parameter regime $4J < \Delta$, and $E_{R,p} = \hbar^2\pi^2/2m_p a^2$. The potential is comblike for $b/a < 1/4$, since the particles resolve the individual molecules forming the crystal, while it is sinusoidal for $b/a \gtrsim 1/4$. The strong dipole-dipole repulsion between the extra particles acts as an effective hard-core constraint [14]. We find that for $4J < \Delta$ and $d_p \ll d_c$ the bare off-site interactions satisfy $V_{ij} \sim d_p^2/(a|i-j|)^3 < \Delta$.

The particle-phonon coupling is $M_q = (d_c d_p/ab) \times (2\hbar/N m_c \omega_q)^{1/2} q^2 \mathcal{K}_1(b|q|)\beta_q$ with $\mathcal{K}_1$ the modified Bessel function of the second kind, and $M_q \sim \sqrt{q}$ for $q \to 0$. For $b/a < 1$, which is the regime of interest (compare Fig. 2), M_q is peaked at large $q \sim \pi/a$, so that the main contribution to the integrals in the definition of S_T and E_p is indeed dominated by large frequencies $\hbar\omega_q > J$. A plot of S_0 as a function of b/a is shown in Fig. 2(a). We find the scaling $S_0 \propto \sqrt{r_d}(d_p/d_c)^2$, and within the regime of validity of the single-band approximation, S_0 can be tuned from $S_0 \ll 1$ ($\tilde{J} \sim J$) to $S_0 \gg 1$ ($\tilde{J} \ll J$) corresponding to the large and small polaron limit, respectively. The polaron shift E_p, as shown in Fig. 2(b), generally exceeds the bare hopping rate J, and, in particular, $E_p \gg J$ for $S_0 \gtrsim 1$. Together with the condition $\hbar\omega_D \gg J$ this ensures that the dynamics of dressed particles is described by Eq. (3) [11].

The phonon-mediated interactions $V_{i,j}^{(1)}$ show oscillations, which for $b/a \lesssim 1/4$ decay slowly as $\sim 1/|i-j|^2$, and are thus long ranged. Depending on their sign, they can enhance or reduce the direct dipole-dipole repulsion of the extra particles. The term $V_{j,j+1}^{(1)}$ alternates between attractive and repulsive as a function of b/a, Fig. 2(c). Figure 2(d) is a contour plot of $\tilde{V}_{j,j+1}/2\tilde{J}$ as a function of r_d and b/a, where $\tilde{V}_{j,j+1}/2\tilde{J}$ increases by decreasing b/a or increasing r_d, and can be much larger than 1.

In *configuration 2* [Fig. 1(c)] neutral atoms are trapped in the same tube as the crystal molecules, and feel the 1D

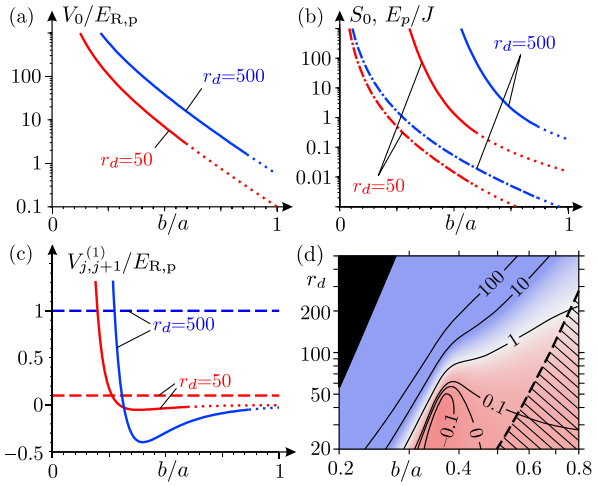

FIG. 2 (color online). Configuration 1 [Fig. 1(b)]: Hubbard parameters for $d_p/d_c = 0.1$ and $m_c = m_p$. (a) Lattice depth V_0 in units of $E_{R,p}$ vs b/a for $r_d = 50$ and 500. Thick continuous lines: tight-binding region $4J < \Delta$. (b) Reduction factor S_0 (dash-dotted lines) and polaron shift E_p/J (solid lines), for $4J < \Delta$. (c) Continuous lines: phonon-mediated interactions $V_{j,j+1}^{(1)}$. Horizontal (dashed) lines: $V_{j,j+1}$. (d) Contour plot of $\tilde{V}_{j,j+1}/2\tilde{J}$ (solid lines) as a function of b/a and r_d. A single-band Hubbard model is valid left of the dashed region ($4\tilde{J}$, $\tilde{V}_{ij} < \Delta$), and right of the black region ($E_p < \Delta$).

static potential $V_{\rm cp}(x) = \sum_j g_{\rm cp}\delta(x - ja)$ (Kronig-Penney model). Here $g_{\rm cp} = 2\hbar^2 a_{\rm cp}/\mu_p a_{\perp,p}^2$ for a 3D scattering length smaller than the transverse confinement, $a_{\rm cp} \ll a_{\perp,p} = (\hbar/m_p\omega_{\perp,p})^{1/2}$, with μ the reduced mass [15]. For $g_{\rm cp}/aE_{R,p} \gg 1$ the width of the lowest band is $4J \simeq (aE_{R,p}/g_{\rm cp})$ and $\Delta \simeq 3E_{R,p}$.

In the following we are interested in bosonic atoms interacting with each other via a contact potential with coupling strength $g_{\rm pp}$ determined by their 3D scattering length $a_{\rm pp}$, which is tunable independent of $a_{\rm cp}$. The bare Hubbard interactions are dominated by on-site interactions, $V_{i,j} \simeq \delta_{ij} U$, where $U = 3g_{\rm pp}/2a$ for large $g_{\rm cp}$. For the parameters of Fig. 3, a single-band Hubbard model is valid for $g_{\rm cp}/aE_{R,p} \gtrsim 0.5$ so that $J, U \ll \Delta$.

The particle-phonon coupling is $M_q = (g_{\rm cp}/a) \times (2\hbar/N m_c \omega_q)^{1/2}|q|\beta_q$, which is peaked at large $q \sim \pi/a$, and $M_q \sim \sqrt{q}$ for $q \to 0$. We find that $S_0 \sim 0.92(g_{\rm cp}/aE_{R,p})^2 (m_c/m_p)^2/r_d^{3/2}$ and $E_p \sim 0.94E_{R,p}(g_{\rm cp}/aE_{R,p})^2(m_c/m_p)/r_d$ *decrease* with increasing r_d [see Fig. 3(a)]. For the parameters of Fig. 3 we have $S_0 \ll 1$ over a wide range of r_d, so that $\tilde{J} \approx J$. The strong coupling regime $S_0 \gg 1$, with $E_p/J \gg 1$, can be reached by decreasing r_d or increasing $g_{\rm cp}/aE_{R,p}$. We note that $g_{\rm cp}/aE_{R,p}$ is restricted by the condition of a stable crystal $\Delta u < a$, i.e., $g_{\rm cp}/aE_{R,p} < 3r_d(1 - 0.4/r_d^{1/4})$.

While the bare atom-atom interaction provides only an on-site shift, the phonon coupling induces long-range in-

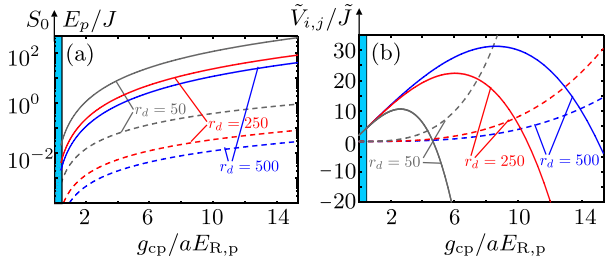

FIG. 3 (color online). Configuration 2 [Fig. 1(c)]: Hubbard parameters for $m_p/m_c = 0.814$, $a_{pp} = 2$ nm, and $\omega_{\perp,c}/2\pi = 200$ kHz. (a) Reduction factor S_0 (dashed lines) and polaron shift E_p/J (solid lines) as a function of the particle-phonon coupling strength $g/aE_{R,p}$. The tight-binding region, $4J, V_{j,j} < \Delta$, is right of the shaded area. (b) Effective on-site $\tilde{V}_{j,j}/\tilde{J}$ (solid lines) and nearest-neighbor $\tilde{V}_{j,j+1}/\tilde{J}$ (dashed lines) parameters.

teractions $V^{(1)}_{i,j}$, which decay as $\sim 1/|i-j|^2$. The effective interactions $\tilde{V}_{j,j}$, $\tilde{V}_{j,,j+1}$, and $\tilde{J}$ are summarized in Fig. 3(b), where the nearest-neighbor term $\tilde{V}_{j,j+1} \sim 0.16E_p$ is shown to be repulsive for all values of $g_{cp}/aE_{R,p}$, while the on-site interaction $\tilde{V}_{j,j} = U - 2E_p$ turns from positive to negative, which for bosons indicates an instability towards collapse. That is, for stability we require $g_{pp}/aE_{R,p} \gtrsim 1.4(g_{cp}/aE_{R,p})^2(m_c/m_p)^2/r_d$. Strong interactions $\tilde{V}_{j,j}, \tilde{V}_{j,j+1} \gg \tilde{J}$ are reached for $r_d \gg 1$, Fig. 3(b).

One feature of our extended Hubbard model is the appearance and tunability of strong off-site interactions, a necessary ingredient for a variety of new quantum phases [16,17], which is difficult to realize in a standard atomic setup [18]. As an example, at half filling the particles in configuration 1 undergo a transition from a (Luttinger) liquid ($\tilde{V}_{i,i+1} < 2\tilde{J}$) to a charge-density wave (CDW) ($\tilde{V}_{i,i+1} > 2\tilde{J}$), which can be observed, e.g., for $r_d = 100$ at $b/a \approx 0.5$ [see Fig. 2(d)] [19]. Similarly, the ground state of configuration 2 at half filling is a CDW for $\tilde{V}_{j,j}, \tilde{V}_{j,j+1} > 4\tilde{J}$ [19]. For larger filling and strong interactions $2\tilde{V}_{j,j+1} \gtrsim \tilde{V}_{j,j} > 4\tilde{J}$, a second order transition occurs to a supersolid phase, where diagonal and off-diagonal orders coexist [20]. These strong interactions are here realized, e.g., for $r_d = 250$ at $g_{cp}/aE_{R,p} \approx 10$, where $\tilde{V}_{j,j+1} \approx V_{j,j} \approx 7.6\tilde{J}$ [see Fig. 3(b)].

In conclusion, we have studied a scenario where cold atoms or molecules move in the periodic potential provided by a dipolar molecular crystal, with quantum dynamics given by phonons. Strong phonon-mediated off-site interactions and particle localization open new perspectives for studying the interplay between strong correlations and phonon dynamics in a tunable setup.

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