Quantum dimer models— which describe the dynamics of close-packed hard-core dimers on a lattice— have received continued attention since their original proposal by Rokhsar and Kivelson in 1988 [1]. Several factors have motivated these studies, including connections to Pauling and Anderson resonating valence bonds [2, 3], the appearance of quantum critical points [4, 5], topological order and fractionalized excitations [6–8], their mapping to lattice gauge theories [9–13], and their potential applications in quantum computation [14–16]. This rich variety of physics emerges due to the interplay between quantum fluctuations, hard-core constraints, and the lattice geometry of these systems. There are relatively few experimental realizations of dimer models. In this Letter, we show that experiments using an atomic gas trapped in an optical lattice can realize and probe subtle dimer resonances in a range of quantum dimer models.

Our technique works for atoms (fermionic or bosonic), with electronic orbital angular momentum, \( l = 0 \), relatively large hyperfine spin \( f \), but relatively weak dipole-dipole interactions. Alkalis such as \( ^{87}\text{Rb} \) with \( f = 2 \), or \( ^{40}\text{K} \) with \( f = 9/2 \) are ideal. By tuning the lattice depth and trapping potential one drives the system into a Mott insulating state with one atom per site [17]— effectively yielding immobile spins on each site. These spins interact via a virtual superexchange process [20]. We propose manipulating these superexchange interactions by optically coupling pairs of atoms to an excited molecular state which has \( S = 1 \) and \( L = 0 \). When tuned sufficiently off-resonance, this optical coupling favors the formation of nearest-neighbor hyperfine singlets, which we refer to as dimers. In the large-\( f \) limit the dimers are monogamous and orthogonal, e.g., a state where site \( i \) forms a singlet with site \( j \) is orthogonal to one in which \( i \) forms a singlet with \( k \neq j \). The resulting theory has the form of a quantum dimer model and, depending on lattice geometry and scattering length parameters, has the potential to realize dimer crystals and dimer liquid (resonating valence bond) ground states.

There have been previous proposals to observe related physics in cold atom experiments, including crystallized dimer phases [21, 22], resonating plaquette phases [23, 24], and dimer liquid phases [25–27]— all hallmarks of the quantum dimer model. However, these studies were generally based on different mechanisms, and did not exploit mappings of their systems onto quantum dimer models.

Here, we explicitly show that quantum dimer models describe the effective low-energy interactions of large-spin Mott insulating atoms in a unit-filled optical lattice, when the effective nearest-neighbor interactions are dominated by scattering processes through the spin-singlet channel. In the simplest case, we find the columnar dimer state on the square lattice and the \( \sqrt{12} \times \sqrt{12} \) plaquette phase on the triangular lattice as in Ref. [28]. The ground states for the 2D square and 3D cubic lattices are in general a matter of contention [29–33], which an experimental realization of our proposal could resolve. Furthermore, we present protocols to detect both dimer crystal phases and dimer liquid phases using quantum gas microscopy and photoassociation. We explain how to further tune parameters to explore phase space where a dimer liquid phase may be stabilized.

Microscopic model— To produce a quantum dimer model, we begin with a tight-binding Hamiltonian for atoms in an optical lattice, which includes spin-dependent interactions. We obtain this Hamiltonian from the continuum theory (e.g., Ref. [37]) by transforming to the Wannier...
than all others: significantly weaker in the hyperfine singlet channel in Feshbach resonance so that the interactions are similar magnitude, we argue that one can engineer an optical model.

On bipartite lattices, this model is an example of an $SU(N)$ antiferromagnet model. We show below that our model [Eq. (3)] can be mapped onto a dimer model.

For typical parameters (lattice depth $V_x = V_y = 11E_R$, $V_z = 50E_R$, wavelength $\lambda = 532$ nm, and scattering length $a_0 = 50$ Bohr), the superexchange coefficient is $2J^2/U_0 = 10.5$ Hz. This scale is large compared to neglected physics such as off-site dipole interactions ($\sim 0.7$ Hz for alkali atoms). One may also worry about tensor light shifts from the lattice or photo-association beams. These will be minimal if the laser detunings are larger than hyperfine splitting [10]. If any residual light shifts remain, they can be cancelled by adding additional fields (e.g. as in Ref. [11]).

**Tuning the interactions via an Optical Feshbach Resonance**—We induce an optical Feshbach resonance [12–51] between pairs of atoms by shining a laser tuned near a transition to an excited molecular state, labeled by orbital angular momentum $L = 1$, electronic spin $S = 0$, and total electronic angular momentum $J_e = 1$. The nuclear angular momentum is not important as long as the detuning of the laser is large compared to the hyperfine splitting. In the cold collision limit the rotational angular momentum of the nuclei vanishes, $R = 0$. As in Ref. [52], second order perturbation theory then gives a contribution to $U_F = U_F^{bg} + U_F^{Fesh}$ of

$$U_F^{Fesh} = \alpha_F \frac{\Omega^2}{\delta + i\Gamma/2},$$

with $U_F^{bg}$ encoding the background scattering, including any influence of on-site dipole-dipole interactions. The matrix element $\Omega^2$ is proportional to the intensity of the laser. The detuning $\delta$ is typically much larger than the linewidth $\Gamma$ ($\sim 100$ MHz), so molecular decay can be neglected [43]. The coefficient $\alpha_F$ is the square of the overlap between the electronic spin singlet ($S = 0$) and the hyperfine state with total spin $F$. It is related to the Wigner $9j$ symbols. As detailed in the Supplementary Material, for alkali atoms, which have $s = 1/2$ and nuclear spin $I > 0$, we calculate $\alpha_F = 2(f+1)(f+2/F)(F+1/2)/(2(f+1/2)^2)$.

Assuming that $U_F^{bg}$ depends only weakly on $F$, one can then choose the laser intensity and detuning so that $U_F \neq 0 \Rightarrow U_0 \gg |U_F^{Fesh}|$, which then yields Eq. (3).

**From Singlet Coverings to Dimer Models**—The low energy eigenstates of Eq. (3) can be written as a superposition of singlet coverings, with each singlet cover being constructed from the pair operators as follows:

$$|a\rangle = \prod_{(i,j) \in a} \hat{A}^+_{ij} |0\rangle,$$

where $(i,j)$ refers to a singlet bond between sites $i$ and $j$, which are not necessarily nearest neighbors. In Eq. (5), each lattice site appears exactly once in the product over bonds. Some examples of singlet coverings are illustrated in Fig. 1. As explicitly shown in the supplementary materials, this subspace of states is closed under the operation of $\hat{H}_{\text{eff}}$.

The singlet coverings defined in Eq. (5) are not orthogonal, and their overlap $S_{a,b} = \langle a | b \rangle$ may be calculated through their transition graph, as shown in Fig. ?? of the supplementary materials. We follow Ref. [11] to construct an orthogonal basis $|\bar{a}\rangle$ from these non-orthogonal singlet coverings, for which $\langle \bar{a} | \bar{a}' \rangle = \delta_{a,a'}$ and

$$|\bar{a}\rangle = \sum_b \sqrt{S^{-1}_{a,b}} |b\rangle.$$

We call these orthogonal basis states $|\bar{a}\rangle$ dimer states. At large $f$ the singlet coverings are nearly equal to their
associated dimer states and so effectively become orthogonal as well.

If we take $f \to \infty$, the dimer covers $|\tilde{a}\rangle$ are eigenstates of $H_{\text{eff}}$ with energy $E_a = \frac{-2J}{a} N_a$, where $N_a$ is the number of nearest neighbor bonds in $|\tilde{a}\rangle$. In this limit, the states with only nearest-neighbor bonds span the degenerate ground state manifold. We then use second-order degenerate perturbation theory in $(2f + 1)^{-1}$ to calculate an effective model in this subspace. On a square, triangular, or a cubic lattice, we find

$$H_{\text{QDM}} = \sum t |i\rangle \langle i| + \text{H.c.} + V |i\rangle \langle i| + |i\rangle \langle i|$$

$$- t' |i\rangle \langle i| + \text{H.c.}. \quad (7)$$

The first term in this effective theory—parameterized by $t$—models a rotation of two parallel bonds within a four-site plaquette from a vertical to a horizontal configuration, or vice versa. Such plaquettes containing two parallel bonds are called flippable plaquettes. The second term—parameterized by $V$—models an effective interaction between two parallel bonds inside a flippable plaquette. The third term—parameterized by $t'$—models a resonance for three bonds within a six-site plaquette. The effective dimer model on the honeycomb and Kagome lattices have similar terms, but the smallest kinetic term involves three-bond loops, and therefore $t$ has an amplitude of $O(2f + 1)^{-2}$. Table I lists the parameters $t, t'$ and $V$ for different lattice geometries.

Our proposal to produce a quantum dimer model using cold atoms has several advantages over electronic systems. First, using large spin atoms allows for a controlled expansion in terms of the small parameter $(2f + 1)^{-1}$. This is in contrast to spin-1/2 electronic systems where orthogonality issues are neglected and expansions are often truncated without a small parameter. Second, moving between optical lattice geometries is relatively easy in systems with cold atoms [34,35], allowing for straightforward exploration of the phase diagram for this model. Third, dimer models studied in electronic systems typically have $t' = 0$; our proposal produces a richer model with $t' \neq 0$.

**Phases** — The effective model for our system from Eq. (7) has a rich phase diagram, which has been well explored along $t' = 0$ in a number of geometries [6,11,15,16,34,36,37,38]. For 2D bipartite lattices with $t' = 0$, one finds only valence bond solid phases, while on 3D and non-bipartite 2D lattices dimer liquids may be found. The phase diagram at finite $t'$ is less explored [39].

The valence bond solid phases described in the literature fall into four types: columnar, plaquette, mixed, and staggered. The columnar phase is built from vertical columns of horizontal parallel bonds, or vice versa. In the plaquette phase, dimer bonds resonate between different configurations inside a multi-site unit cell. For example, on a square lattice, the plaquette phase has a unit cell with four lattice sites; two parallel bonds resonate between horizontal and vertical configurations inside a plaquette. The plaquette phases on a triangular lattice have larger unit cells. The mixed phase is a hybrid between the columnar and plaquette phases. The staggered phase has no flippable plaquettes. The columnar phase is favored at large negative $V$, and the staggered phase at large positive $V$.

In nearly all theoretical analyses, our model on a square lattice yields a columnar phase as $f \to \infty$. There is some contention about the presence of plaquette and mixed phases as the ground state on a square lattice, and the location of the phase boundaries [36], which could be resolved through experiments. On the triangular lattice, as $f \to \infty$, we expect to see a plaquette phase, called the $\sqrt{12} \times \sqrt{12}$ phase, which has a 12-site unit cell, and quantum resonances that extend throughout the cell [34]. Observing these resonances is part of Pauling and Anderson’s vision of quantum resonances that manifest throughout a macroscopic system [3].

By tuning the Feshbach parameters, our proposal to produce the dimer model can be extended to explore even more exotic regions of the model’s phase diagram — for

![FIG. 1. (Color online) Examples of singlet cover states. The numbers label the lattice sites, while connected sites represent a spin singlet between the atoms on those sites. In this example, $|a\rangle = A_{1,2}^{1,2}A_{1,5}^{1,5}A_{1,9}^{1,9} |0\rangle$. The notation $|(i,j) : a\rangle$ denotes a state where sites $i$ and $j$ are paired in a singlet, the original partners of $i$ and $j$ in $|a\rangle$ are paired in another singlet, and all the other bonds in $|a\rangle$ are left unchanged. This notation is used in the supplementary materials to derive an effective dimer model [Eq. (7)].](image-url)
example, bringing one to the highly degenerate Rokhsar-Kivelson point or producing a spin liquid. If we tune the coupling and detuning such that $U_{F>0.2} > U_2 > U_0 \gg J$, then the effective Hamiltonian at second order in the tunneling strength is

$$\hat{H}_{\text{eff}} = -\frac{2J^2}{U_0} \sum_{(ij)} \hat{A}_{ij}^{00\dagger} \hat{A}_{ij}^{00} - \frac{2J^2}{U_2} \sum_{(ij)M} \hat{A}_{ij}^{2M\dagger} \hat{A}_{ij}^{2M}, \quad (8)$$

where $\hat{A}_{ij}^{2M\dagger} = \sum_m \hat{C}_{m,M-M}^{2M} \hat{b}_{ij} \hat{b}_{ij}$ creates a neighboring atom pair with total spin $F = 2$ and azimuthal spin $M$. By applying the same large-$f$ perturbation techniques, we obtain a dimer model [Eq. (7)], where $V$ and $t'$ depend on $f$, $J^2/U_0$, and $J^2/U_2$. The amplitude $t$ develops corrections at $O\left(f^{-2}\right)$. By tuning $U_2/U_0$ via the Feshbach laser intensity and detuning, one can control these parameters.

Detection—To probe the valence bond solid order and observe the resonating dimers in the plaquette and spin liquid phase, we propose measuring the dimer-dimer correlation function $\langle \hat{A}_{ij}^\dagger \hat{A}_{kl}^\dagger \hat{A}_{kl} \hat{A}_{ij} \rangle$. We provide an experimental protocol to image dimer bonds and measure dimer-dimer correlations, which researchers have calculated for quantum dimer models with $t' = 0$ for different lattice geometries [10, 24, 35, 60]. Furthermore, we numerically calculate these correlations in our system for both large and moderate values of $f$, thereby characterizing the ground state of our model.

To image the dimer bonds we propose shining a weak near-resonant photoassociation laser on the system, tuned near a molecular state with angular momenta $L = 1$ and $S = 0$. Then, when virtual hopping brings two atoms forming an $S = 0$, $L = 0$ dimer onto the same lattice site, the near-resonant light drives these atoms into the molecular state. The excited molecule has a short lifetime and so those atoms are lost from the trap. The probability of losing adjacent atoms which do not form a dimer is proportional to $1/(2J^2 + J^2)$. Subsequently we use a quantum gas microscope to image the location of all remaining atoms [64, 65]. All adjacent pairs of empty sites in the image were likely occupied by atoms entangled in dimers. In this way, a fraction of the dimers in the system can be imaged. Dimer-dimer correlations can be extracted by analyzing data from multiple realizations of this imaging process, and can be used to identify the phase. The ability to map out the correlations in a quantum dimer model will allow an unambiguous identification of the phases and their order.

To find out what these experiments may reveal, we simulate our system numerically by computing the ground state of the model in Eq. (3). The natural basis for a numerical simulation is spanned by the non-orthogonal singlet coverings. In our numerics, we do not rely on the large-$f$ approximation, nor do we restrict ourselves to the low-energy sector which contains only states with nearest-neighbor dimer bonds. However, we are limited to a small system with no more than 16 lattice sites. By exactly diagonalizing the Hamiltonian of Eq. (3), we compute the ground state and the dimer-dimer correlation functions $\langle \hat{A}_{ij}^\dagger \hat{A}_{ij}^\dagger \hat{A}_{kl} \hat{A}_{kl} \rangle$ for both square and triangular lattices.

Figure 2 shows the calculated correlations between a bond $(i,j)$ and a bond $(k,l)$ on a square lattice for two different values of $f$, where the bond $(i,j)$ is fixed, and the bond $(k,l)$ is varied. For large $f$ we find correlations

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure2}
\caption{Dimer-dimer correlations on a square lattice with periodic boundary conditions, for (a) $f = 100$ and (b) $f = 3$. The reference dimer is marked grey in the lower left corner. The thickness of the lines is proportional to the strength of correlation, which is indicated on the lines.}
\end{figure}

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure3}
\caption{Dimer-dimer correlations on a triangular lattice for (a) $f = 100$ and (b) $f = 3$. The reference dimer is marked in grey. The thickness of the lines is proportional to the strength of correlation.}
\end{figure}
consistent with a columnar phase. For \( f = 3 \), the correlations are weaker but still indicative of a columnar phase. Figure 4 shows the correlations on a triangular lattice for the same two values of \( f \). For this lattice, we anticipate the ground state to be the \( \sqrt{12} \times \sqrt{12} \) phase [34], and so we take our system to have the shape of a unit cell in the \( \sqrt{12} \times \sqrt{12} \) phase. For both large and moderate values of \( f \), we find correlations consistent with the \( \sqrt{12} \times \sqrt{12} \) phase.

We also use these numerical results to investigate the stability of our system against small perturbations, such as off-site dipole-dipole interactions. We calculate the matrix element of this perturbation \( V_{01} \) between the grounds state and first excited state. Using parameters appropriate for Rubidium, we find \( V_{01} \sim 0.008 \text{ Hz} \), which is small compared to the lowest energy plaquette-flip excitation \( \sim 0.9J^2/U_0 \sim 4.5 \text{ Hz} \).

We emphasize that the ability to directly image the valence-bond correlations is one of the greatest strengths of this system, and will allow unambiguous identification of the various valence-bond ordered phases. Spin liquid phases will be characterized by the absence of long-range valence bond order.

Summary — In summary, we propose experimental protocols to produce quantum dimer models and detect both static and resonating patterns of dimer configurations. In particular, we show that appropriately tuned off-resonant photoassociation light modifies the interactions in a gas of cold atoms, yielding a low energy Hilbert space spanned by short-range dimers. We develop an effective dimer model Hamiltonian in powers of \((2f+1)^{-1}\), and discuss its phase diagram. We find that a number of valence bond solid and plaquette phases are readily produced, and suggest techniques which are suited to searching for even more exotic states such as topological spin liquids. We demonstrate that by combining photoassociation with quantum gas microscopy one can directly detect the dimers and the dimer-dimer correlations, thereby probing the defining features of these phases. We numerically calculate the dimer correlations, finding that on triangular lattices one will be able to image an intricate pattern of resonating bonds, extending over a 12-site unit cell.

Quantum dimer models have been highly influential in developing an understanding of how geometric constraints lead to new emergent physics [59, 60], and have been used as a theoretical foundation for attempting to understand phenomena ranging from high temperature superconductivity to exotic antiferromagnets [67]. A direct experimental realization of dimer models is key to validating and refining these ideas.

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SUPPLEMENTARY MATERIALS

CALCULATION OF INTERACTION COEFFICIENTS $\alpha_F$ IN DIFFERENT SPIN CHANNELS

Here we calculate the coefficients $\alpha_F$ that parameterize the interactions $U_F$ in the different spin channels. When we shine a laser tuned near a molecule with $L = 1$ and $S = 0$, with a detuning large compared to hyperfine coupling, the effective interaction between atoms from second order perturbation theory is

$$\hat{H}_{\text{int}} = \frac{\Omega^2}{\delta + i\Gamma/2} \sum_{j,m,m'} \left( \frac{\hat{b}_{j\uparrow m} \hat{b}_{j\downarrow m'} - \hat{b}_{j\downarrow m} \hat{b}_{j\uparrow m'}}{\sqrt{2}} \right) \left( \frac{\hat{b}_{j\downarrow m'} \hat{b}_{j\uparrow m} - \hat{b}_{j\uparrow m} \hat{b}_{j\downarrow m'}}{\sqrt{2}} \right), \quad (S1)$$

where $\hat{b}_{j\sigma m}$ annihilates a particle on lattice site $j$ with electronic spin projection $\sigma = \uparrow, \downarrow$ and nuclear spin projection $m$. This interaction can be written in the total spin channels as

$$\hat{H}_{\text{int}} = \frac{\Omega^2}{\delta + i\Gamma/2} \sum_{F,M} \alpha_F \hat{A}_{jF}^M \hat{A}_{jF}^M, \quad (S2)$$

where $\alpha_F$ is independent of $M$ due to $SO(3)$ spin rotation symmetry, and $\hat{A}_{jF}^M$ were defined in Eq. (2). Since the interaction is local, we drop the spatial index from the operators in this section. We find from Eqs. (S1) and (S2) that

$$\alpha_F = \langle 0 | \hat{A}^M \sum_{m,m'} \left( \frac{\hat{b}_{j\uparrow m} \hat{b}_{j\downarrow m'} - \hat{b}_{j\downarrow m} \hat{b}_{j\uparrow m'}}{\sqrt{2}} \right) \left( \frac{\hat{b}_{j\downarrow m'} \hat{b}_{j\uparrow m} - \hat{b}_{j\uparrow m} \hat{b}_{j\downarrow m'}}{\sqrt{2}} \right) \hat{A}_{jF}^M | 0 \rangle. \quad (S3)$$

We choose to calculate the right hand side of Eq. (S3) for $M = 0$. To evaluate this expression, we project the operators $\hat{b}_{\sigma m}$ into the hyperfine manifolds using the relation

$$\hat{b}_{\sigma m} = \sigma \sqrt{f + 1/2 - \sigma m} \hat{b}_{f\,i=\downarrow} - \sigma \sqrt{f + 1/2 + \sigma m} \hat{b}_{f\,i=\uparrow}, \quad (S4)$$

where $\sigma = +1(-1)$ corresponds to $\uparrow(\downarrow)$, and the operators $\hat{b}_{f\,i=\pm\uparrow/2}$ annihilate particles with hyperfine spin $n$ in the $f = i \pm 1/2$ manifold. One of these is the ground state manifold, and the other an excited manifold. In our manuscript, we denote the operators in the ground state manifold as simply $\hat{b}_n$. We obtain a closed form expression for $\alpha_F$ from Eqs. (S3) and (S4):

$$\alpha_F = \frac{1}{2(i + 1/2)} \sum_n C_{n,-n}^{F_0} \left( C_{n,-n}^{F_0} (n^2 + (f + 1)^2) - C_{n+1,-n-1}^{F_0} ((f + 1/2)^2 - (n + 1/2)^2) \right), \quad (S5)$$

where $C_{n,-n}^{F_0} = \langle f, n; f, -n|F, 0 \rangle$ is a Clebsch-Gordan coefficient. Using the identities

$$\sum_n C_{n,-n}^{F_0} C_{n,-n}^{F_0} = 1, \quad (S6)$$

$$\sum_n C_{n,-n}^{F_0} C_{n,-n}^{F_0} n^2 = \langle F, 0 | -\hat{f}_1 \hat{f}_2 | F, 0 \rangle,$$

$$\sum_n C_{n,-n}^{F_0} C_{n+1,-n-1}^{F_0} ((f + 1/2)^2 - (n + 1/2)^2) = \langle F, 0 | \hat{f}_1 \cdot \hat{f}_2 - \hat{f}_1 \hat{f}_2 | F, 0 \rangle,$$

$$\langle F, 0 | \hat{f}_1 \cdot \hat{f}_2 | F, 0 \rangle = \frac{F(F+1)}{2} - f(f+1),$$

we reduce Eq. (S5) to a simple expression for $\alpha_F$:

$$\alpha_F = \frac{(f+1)(2f+1) - F(F+1)/2}{2(i + 1/2)^2}. \quad (S7)$$
FIG. S1. (Color online) Examples of transition graphs between non-orthogonal singlet coverings. The magnitude of the overlap is given by Eq. (S8) and is shown in the figure for the two cases. The overlap \( S_{ab} \) comes from a single 4-site loop, and represents the largest possible overlap in magnitude. The overlap \( S_{ac} \) comes from a single 6-site loop, and is down in magnitude by a factor of \( (2f+1)^{-1} \). In the large-\( f \) limit, all singlet coverings become orthogonal as the overlaps approach zero.

CONSTRUCTION OF AN ORTHOGONAL BASIS

Here we construct an orthogonal basis from the non-orthogonal singlet coverings, perturbatively in \( (2f+1)^{-1} \). The singlet coverings contain both short- and long-ranged singlet bonds. We use the commutation relation

\[
\left[ \hat{A}_{ij}, \hat{A}_{ij}' \right] = \delta_{jj'} \left( 1 + \frac{\hat{n}_i + \hat{n}_j}{2f+1} \right) + \frac{1 - \delta_{jj'}}{2f+1} \sum_m \hat{b}_{j',m} \hat{b}_{j,m},
\]

to calculate the overlap between states. We find that the overlap \( S_{ab} = \langle a | b \rangle \) between two singlet coverings \( |a\rangle \) and \( |b\rangle \) is

\[
S_{ab} = (2f+1)^{-N_{\text{loops}} N_{\text{loops}}} \left( \frac{1}{\sqrt{2f+1}} \right)^{L_{\text{loops}}}, \tag{S8}
\]

where \( N_{\text{loops}} \) is the total number of closed loops formed by the dimers not common to \( |a\rangle \) and \( |b\rangle \), while \( L_{\text{loops}} \) is the total number of sites involved in all loops. This overlap matrix element is pictorially represented by a transition graph, shown in Fig. S1. For large \( f \), we expand \( S_{ab} \) in powers of \( (2f+1)^{-1} \) to obtain

\[
S_{ab} = \delta_{ab} + \frac{\Box_{ab}}{2f+1} + \frac{\Box_{ab}^{(2)}}{(2f+1)^2} + \mathcal{O}(f^{-3}). \tag{S9}
\]

Here, \( \Box_{ab} = 1 \) if \( |a\rangle \) and \( |b\rangle \) differ by a 4-site loop in their transition graph, and is zero otherwise. The symbol \( \Box_{ab}^{(2)} = 1 \) if \( |a\rangle \) and \( |b\rangle \) differ by either a single 6-site loop, or two distinct 4-site loops in their transition graph, and is zero otherwise. The expression for the orthogonal dimer states from Eq. (6) in the main text then reduces to

\[
|\bar{a}\rangle = |a\rangle - \sum_b \left( \frac{\Box_{ab}}{2(2f+1)} + \frac{\Box_{ab}^{(2)}}{2(2f+1)^2} - \frac{3}{8(2f+1)^2} \sum_c \Box_{ac} \Box_{cb} \right) |b\rangle + \mathcal{O} \left( f^{-3} \right). \tag{S10}
\]

We note that a given dimer state contains an \( \mathcal{O}(1) \) contribution from a particular singlet covering, which we may use to label the dimer state. Higher-order contributions come from singlet covers which differ from the labeling cover by successively longer loops in their transition graph. A pictorial example of singlet covers being used to construct an orthogonal dimer state is shown to first order in Fig. S2.

DERIVING AN EFFECTIVE DIMER MODEL

The Hamiltonian [Eq. (3)] contains terms \( \hat{A}_{ij}^{\dagger} \hat{A}_{ij} \) which are restricted to nearest-neighbor pairs \( \langle ij \rangle \) only. The action of one of these terms on a singlet covering \( |a\rangle \) is given by

\[
\hat{A}_{ij}^{\dagger} \hat{A}_{ij} |a\rangle = \begin{cases} |a\rangle, & \text{for } (i,j) \in a \\ (2f+1)^{-1} |(i,j) : a\rangle, & \text{for } (i,j) \notin a, \end{cases} \tag{S11}
\]
where the notation \( |(i, j) : a\rangle \) denotes a state where sites \( i \) and \( j \) are paired together into a singlet, the original partners of \( i \) and \( j \) in \(|a\rangle\) are paired together into a singlet, and all the other bonds in \(|a\rangle\) are left unchanged. An example of a singlet covering \(|a\rangle\) and a few of the related states \(|(i, j) : a\rangle\) are illustrated in Fig. 1 of the main text.

To derive an effective model in the Hilbert space spanned by dimer states, we first trivially rewrite the Hamiltonian in Eq. (3) as

\[
\hat{H} = -\frac{1}{2(2f+1)} \left( \sum_{(ij)} A_{ij} \hat{A}_{ij} - \hat{N}_{nn} \right),
\]

where \( \hat{N}_{nn} \) counts the number of dimers which connect nearest-neighbor sites. The first term, \( -\hat{N}_{nn} \), is diagonal in the dimer basis. The low-energy eigenspace of \( -\hat{N}_{nn} \) is spanned by the set of all nearest-neighbor-only singlet coverings, which have a degenerate eigenvalue of \(-N/2\), where \( N \) is the number of lattice sites. This low-energy eigenspace is separated from higher-energy subspaces by at least \( 2f^2/U_0 \). The second term in Eq. (12) is our perturbation, \( \hat{V} = -\left( \sum_{(ij)} \hat{A}_{ij} \hat{A}_{ij} - \hat{N}_{nn} \right) \), which has matrix elements of \( O(f^{-1}) \). We use degenerate second-order perturbation theory to expand \( \hat{V} \) as a series in \((2f+1)^{-1}\), restricted to the low-energy subspace of nearest-neighbor-only dimer coverings:

\[
\hat{V}_{\text{eff}} = \sum_{ab} |\bar{a}\rangle \langle \bar{b}| \left( \langle \bar{a} | \hat{V} | \bar{b} \rangle + \sum_c \left( \frac{\hat{V}}{\hat{N}_{nn} - N/2} \right) |\bar{c}\rangle \langle \bar{c}| \right).
\]

Here, \(|\bar{a}\rangle \) and \(|\bar{b}\rangle \) lie in the nearest-neighbor-only subspace, while \(|\bar{c}\rangle \) lies in the higher-energy subspace, and contains at least one long-ranged dimer. The matrix element for \( \hat{V} \) between any two dimer states \(|\bar{a}\rangle \) and \(|\bar{b}\rangle \) is given by

\[
\langle \bar{a} | \hat{V} | \bar{b} \rangle = N_b \delta_{ab} - \sum_{cd} \left( \sqrt{S^{-1}} \right)_{ac} \left( N_d S_{cd} + \frac{1}{2f+1} \sum_{(ij) \notin d} S_{c,(ij)d} \right) \left( \sqrt{S^{-1}} \right)_{db},
\]

where \( c \) and \( d \) include all dimer states, and \( (ij) \notin d \) indicates a sum over all nearest-neighbor pairs from state \( d \) which do not together form a dimer.

Substituting Eqs. (9) and (14) into Eq. (13), we find that

\[
\hat{V}_{\text{eff}} = \sum_{ab} |\bar{a}\rangle \langle \bar{b}| \left( -\frac{1}{2f+1} \sum_{(ij) \notin b} \delta_{a,(ij):b} - \frac{1}{(2f+1)^2} \sum_e \sum_{(ij) \notin e} \sum_{(i') \notin (i')b} \delta_{a,(ij):e} \delta_{a,(i')b} \right).
\]

As it stands, this result is independent of the lattice geometry. To simplify further, and to determine the coefficients \( t, t', \) and \( v \) which appear in Table I, we evaluate this expression for different lattices. Here we focus on a square lattice, but the perturbing term \( \hat{V}_{\text{eff}} \) can be reduced to a simple form on other lattices as well. On a square lattice we find

\[
\hat{V}_{\text{eff}} = \sum_{ab} |\bar{a}\rangle \langle \bar{b}| \left( -\frac{2}{2f+1} \square_{ab} - \frac{1}{(2f+1)^2} \left( 4\square_{ab} + 2\delta_{ab} (N_{\perp} + N_{\perp} + N_{\perp}) \right) \right).
\]

Here, \( \square_{ab} \) is 1 if \(|a\rangle \) and \(|b\rangle \) differ by a single 6-site loop in their transition graph, and is zero otherwise. \( N_{\perp} \) counts the number of bonds lying end-to-end with a single empty bond between them in \(|a\rangle \), while \( N_{\perp} \) and \( N_{\perp} \) have similar meanings. Observing that

\[
N_{\perp} + N_{\perp} + 2N_{\perp} + 2N_{\perp} = 3N/2,
\]
where $N$ is the total number of lattice sites, we rewrite Eq. (S16) as

$$
\hat{V}_{\text{eff}} = \sum_{ab} |\bar{a}\rangle \langle \bar{b}| \left( -\frac{2}{2f+1} \square_{ab} - \frac{4}{(2f+1)^2} \square\square_{ab} + \frac{4\delta_{ab}}{(2f+1)^2} (N_+ + N_0) - \frac{9N/2}{(2f+1)^2} \right). 
$$

(S17)

The first term—containing $\square_{ab}$—gives the two-bond resonance term with an amplitude $t = \frac{2}{2f+1} \frac{2J^2}{U_0}$ in $\hat{H}_{\text{QDM}}$ [Eq. (7)]. The second term—containing $\square\square_{ab}$—gives the three-bond resonance term with an amplitude $t' = \frac{4}{(2f+1)^2} \frac{2J^2}{U_0}$ in $\hat{H}_{\text{QDM}}$ [Eq. (7)]. While the third term—containing $N_+ + N_0$—gives an effective interaction $V = \frac{4}{(2f+1)^2} \frac{2J^2}{U_0}$ between two parallel bonds around a four-site plaquette in $\hat{H}_{\text{QDM}}$ [Eq. (7)]. The last term is a constant energy shift, which we may ignore.

In summary, the effective dimer model for our system on a square lattice, when written in the common notation, is given by

$$
\hat{H}_{\text{QDM}} = \sum \frac{-4J^2/U_0}{2f+1} (|\rangle \langle | + \text{H.c.}) - \frac{8J^2/U_0}{(2f+1)^2} (|\rangle \langle = | + |\rangle \langle = | - \frac{8J^2/U_0}{(2f+1)^2} (|\rangle \langle = | + \text{H.c.}) .
$$

(S18)

A calculation for other lattice geometries yields a similar effective model, where the amplitudes of the various terms in the model are given in Table I of the main text.