Nanoscale-structured interactions and potential barriers for dipolar quantum gases

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We design dipolar quantum many-body Hamiltonians that will facilitate the realization of exotic quantum phases under the current experimental conditions achieved for polar molecules and magnetic atoms with large dipolar moments. The main idea is to modulate both two-body dipolar interactions and single-body potential barriers on a spatial scale of tens of nanometers to strongly enhance energy scales of engineered many-body systems. This new scheme greatly relaxes the requirement for low temperatures necessary for observing new quantum phases, especially in comparison to Hubbard Hamiltonians for regular optical lattices. For polar molecules, our approach builds on the use of microwave fields to couple rotational energy eigenstates in static electric fields with strong gradients. We illustrate this approach by demonstrating the orientation switching on the nanoscale for the induced electric dipole moment of a polar molecule. This configuration leads to the formation of interface bound states of fermionic molecules with binding energies far exceeding typical energy scales in current experiments. While the concepts are developed for polar molecules, many of the present ideas can be readily carried over to atoms with magnetic dipolar interactions.

I. INTRODUCTION

Recent experimental advances have enabled exploring the physics of dipolar quantum many-body systems in various AMO platforms, with examples including polar molecules [1–16] and magnetic atoms [17–26]. The goal of these experimental efforts is to realize exotic quantum phases of matter, which are predicted to occur due to the long-range character and orientational dependence of dipolar interactions [27]. In particular, ultracold quantum gases loaded into an optical lattice realize generalized Hubbard models, in which off-site interactions can induce supersolidity [28–32]. A major obstacle for the experimental realization of such quantum phases is posed by the smallness of the typical interaction energy scales $E_{\text{int}} \sim d^2/a^3$, where $d$ is the dipole moment and $a$ is the lattice spacing which is set by the optical wavelength $\lambda_L$ of the laser that creates the lattice potential, $a = \lambda_L/2$. These energy scales range from tens of Hz for magnetic atoms [23] to tens of kHz for polar molecules [2, 33, 34], and thus put a stringent requirement on the temperatures $T < E_{\text{int}}$ one has to reach in experiments. The challenge is, therefore, to develop experimental setups in which the typical distance $a$ between particles is much smaller than optical wavelengths, $a \ll \lambda_L$, and in this way boost interaction energy scales and mitigate the requirements on the temperature.

The challenge of increasing relevant energy scales in atomic many-body systems, and thus reducing temperature requirements to observe exotic low-temperature phases can be achieved, for example, in atomic and molecular Hubbard models by designing sub-wavelength optical lattices [35–37], or optical barriers on the scale of tens of nanometers. Recent theoretical proposals [38–41], and first experimental demonstrations [42] have outlined how optical nanoscale barriers for atoms can be obtained in ‘free space’. The underlying physics exploits large gradients of the electric field from laser light in a standing wave configuration on the scale of the optical wavelength. Combined with the nonlinear atomic response of ‘atomic dark states’ this results in free-space sub-wavelength nanobarrers. We note that sub-wavelength
lattices and barriers involve the design of single particle potentials in atomic and molecular many-body systems with spatial resolution on a sub-wavelength scale.

In the present paper we address the problem of designing atomic and molecular many-body systems with long-range dipolar interactions, where both single-body terms (potential barriers) but also two-body (dipolar) interactions are modulated spatially on the ten-nanometer scale. In our work we will mainly focus on the case of polar molecules, and we build on the recent experimental achievements [43] which allow electric DC and AC microwave fields with strong electric field gradients to interact with the electric dipole moment of the polar molecule and drive rotational excitations. Figure 1(a) illustrates the type of many-body problems we are interested in. Here, a proper choice of DC and AC electric fields, and electric field gradients endows the polar molecule with an induced electric dipole moment \( \mathbf{d}(x) \) which along a given spatial direction \( x \) can flip its sign on the nanoscale. A pair of molecules, with the first molecule at \( x > 0 \) and the second molecule at \( x < 0 \) will thus attract each other due to the long-range character of the dipolar interaction, allowing ‘interface bound’ molecular pairs at the bilayer interface \( x = 0 \), while the pair can move freely in the \( x = 0 \) plane, or be confined by additional trapping potentials, e.g., from an optical lattice well. We note that the binding energy of these pairs will be strongly enhanced by the distances of the order of ten nanometers between the polar molecules.

At the same time, this molecular pair will be well protected from close encounter collisions and thus inelastic loss processes. First, dipole moments, and thus dipolar interactions will turn off with one of the molecules approaching \( x = 0 \), i.e., bound molecules will keep a distance defined by the nanoscale variation of the induced dipole moment. Second, molecules on the same side of the interface will, with the dipoles as drawn in Fig. 1(a) pointing in the same direction, repel each other, allowing ‘interface bound’ molecular pairs at the bilayer interface \( x = 0 \), while the pair can move freely in the \( x = 0 \) plane, or be confined by additional trapping potentials, e.g., from an optical lattice well. We note that the binding energy of these pairs will be strongly enhanced by the distances of the order of ten nanometers between the polar molecules.

While the above example illustrates the spatial modulation of two-particle interactions on the nanoscale for polar molecules, nanoscale barriers as single particle potentials, controlling the motion of molecules on the ten nanometer scale, can again be designed with electric field gradients. Such nanoscale barriers can be used to realize double-layer systems as shown in Fig. 2(a). The underlying ideas are similar to those outlined above with optical field gradients with standing light waves. We note, however, that rotational excitations of polar molecules have the unique advantage of long life time relative to electronic excitations as employed in schemes for optical trapping. Finally, while polar molecules with electric dipoles controlled with electric fields promise comparatively strong dipolar interactions, many of the present ideas carry straight over to atoms and magnetic dipolar interactions.

In the following sections, we describe the theory for experimental setups which are behind the nanoscale manipulation of polar molecules and magnetic atoms outlined above. In Sec. II we start with a preview of the Hamiltonians with dipolar interactions and single-particle potential barriers modulated on the nanoscale. Section III discusses the internal level structure of single polar molecules in electric fields and subject to microwave (MW) couplings. This forms the basis of the design of dipolar Hamiltonians. In particular, to engineer nanoscale-structured induced dipolar interactions, we consider the coupling of a pair of rotational states in an effective two-level system (2LS) configuration in Sec. IV. Nanoscale barriers can be realized as we show in Sec. V by coupling three rotational states (three-level system (3LS)) which form an effective Λ-system. We conclude in Sec. VI. Technical derivations are deferred to Appendices A–D.

II. AN OVERVIEW – MODULATING DIPOLAR HAMILTONIANS ON THE NANOSCALE

Before entering the detailed microscopic discussion and derivation of dipolar Hamiltonians modulated on the nanoscale (see Secs. IV and V below), we find it useful to briefly summarize some of the key features underlying the design of modulated two-body dipolar interactions and single-body potential barriers for the case of polar molecules. As outlined in Figs. 1 and 2, to design two-body interactions and single-body terms, we utilize polar
molecules in 2LS and 3LS setups, respectively.

A. Modulating dipolar interactions on the nanoscale in a two-level system setup

For the 2LS setup, let us consider a polar molecule in a spatially inhomogeneous static electric field aligned along the z axis. As shown schematically in Fig. 1(b), the field consists of a strong offset field $\epsilon_0$ and a gradient field $\epsilon' x$. The offset field induces an electric dipole moment which is oriented parallel to $\epsilon_0$, and the magnitude and sign of which depend on the internal state of the molecule. This induced dipole moment couples to the gradient field, and consequently the molecule acquires a state-dependent Stark shift which, depending on the sign of the induced dipole moment, increases linearly along the x axis as illustrated in Fig. 1(c) with dashed lines. By coupling two internal states with opposite dipole moments via a MW field, we obtain dressed states with energies $E_\pm(x)$. We now focus on a molecule in the + channel, for which the position-dependent energy $E_+(x)$ forms an effective trapping potential. As indicated in Fig. 1(c) with orange and blue arrows, and illustrated further in the lower panel of Fig. 1(a), the induced dipole moment $d_{j}^+(x)$ of the molecule changes sign as it moves across the point $x = 0$, where the MW coupling is chosen to be exactly on resonance. The extent of the region in which the MW coupling is resonant and the induced dipole moment changes sign is determined by the gradient of the electric field and the Rabi frequency $\Omega$ of the MW coupling, and can be on the order of tens of nanometers.

Let us now consider two fermionic molecules in the + channel. We assume that the molecules are strongly confined in the z direction. The effective Hamiltonian, which describes the motion of the molecules in xy plane, takes the form

$$H = \sum_{j=1,2} \left[ \frac{p_j^2}{2m} + E_j(x_j) \right] + \frac{d_{j}^+(x_1)d_{j}^+(x_2)}{\rho_1 - \rho_2},$$

where $\rho_j = (x_j, y_j)$ is the position of the $j$th molecule in the xy plane, and $p_j = (p_{x,j}, p_{y,j})$ the corresponding momentum. For simplicity, we ignore here the modification of the dipolar interaction at short distances due to the confinement. Since the dipole moment $d_{j}^+(x_j)$ changes sign when $x_j$ crosses 0, the two molecules attract each other when they are located on different sides of the interface at $x = 0$. If the interaction strength exceeds a threshold value, this gives rise to interface bound states as illustrated in Fig. 1(a) by the grey region which indicates the corresponding fermionic wave function. The Hamiltonian (1) can easily be generalized to the many-body case. This gives rise to a novel class of many-body systems which have two-molecule bound states as an intrinsic feature.

B. Nanoscale potential barriers in a three-level system setup

We now turn to the 3LS setup. Again we consider a spatially inhomogeneous electric field pointing along the z axis. However, in contrast to the 2LS setup, here we assume also the field gradient to be along the z direction, i.e., the field reads $\epsilon_0 + \epsilon' z$. As described above, the strong offset field $\epsilon_0$ induces state-dependent dipole moments which, in turn, couple to the gradient field and thus give rise to position-dependent Stark shifts. We extend the 2LS setup by adding a second MW field to realize an effective $\Lambda$-system as illustrated in Fig. 2(b). The frequencies of the MW fields are chosen such that both couplings are resonant at the same position $z = 0$. This gives rise to two dressed states $E_{\pm}(z)$ with position-dependent energies, and—in analogy to the "dark" state in electronic $\Lambda$-systems—a third state with energy $E_0(z) = 0$ independent of the position. The MW couplings are resonant in a region of size $l$ which is determined by the electric field gradient and the Rabi frequencies $\Omega$ and can, as in the 2LS, be on the order of tens of nanometers. Outside of this region, the zero-energy dressed state corresponds to the same rotational state with induced dipole moment $d_{l}^0$. Inside the resonant region, the zero-energy state is a mixture of the three coupled rotational states. This "twist" of the internal state of a molecule that is moving along the z axis gives rise to an effective potential which takes the form of a repulsive barrier $V_{na}(z)$ of width $l$. As illustrated in Fig. 2(c), the barrier $V_{na}(z)$ can be used to divide a confining potential $V_L(z)$ with characteristic scale $a_L \gg l$ into two sites. If the motion of the molecule is unrestricted in the $xy$ plane, this setup creates a double-layer geometry, where the spacing between the layers is given by $l$.

We now consider two fermionic polar molecules in the double-layer geometry described above. The dipolar interaction of two polar molecules within the same layer is repulsive. For molecules in different layers, the interaction is attractive at short distances when the molecules are in a head-to-tail configuration as illustrated in Fig. 2(a). The effective Hamiltonian for the two molecules reads

$$H = \sum_{j=1,2} \left[ \frac{p_j^2}{2m} + V_0(z_j) \right] + \frac{(d_0^0)^2}{r_{1,2}^2} \left[ 1 - \frac{3(z_1 - z_2)^2}{r_{1,2}^2} \right],$$

where $r_j = (x_j, y_j, z_j)$ and $p_j = (p_{x,j}, p_{y,j}, p_{z,j})$ are, respectively, the position and the momentum of the $j$th molecule, and $r_{1,2} = |r_1 - r_2|$ is the relative distance. $V_0(z) = V_L(z) + V_{na}(z)$ is the effective potential which gives rise to the double-layer geometry. Also in this setup, the attractive interaction between molecules in different layers can lead to the formation of bound states for sufficiently strong dipolar interactions. In a dipolar many-body system of fermionic polar molecules, such attractive interactions can induce inter-layer pairing [44]
III. MICROSCOPIC MOLECULAR HAMILTONIAN

We proceed with a detailed microscopic derivation of the dipolar Hamiltonians outlined in the previous section. A first goal of the following is to engineer many-body models for polar molecules with dipolar interactions, where the (induced) dipole moment of a molecule depends on its position in space. We note that the dipole moment is determined by the internal state of a molecule. Consequently, we are seeking a situation in which the internal state of a molecule adiabatically follows its motion. This can be achieved due to the separation of scales between the energies associated with the motional degrees of freedom and typical excitation energies of rotational states of diatomic molecules, which leads to the occurrence of approximately decoupled Born-Oppenheimer (BO) channels. As we show below, versatile tools to design such channels are spatially inhomogeneous electric fields and MW couplings. However, to prepare the ground, in this section we first discuss modifications of the rotational level structure in static electric fields, and then how the internal states of molecules in static fields can be coupled by MW radiation.

A. Rigid rotor and Stark effect

The rotational level structure of a diatomic molecule is described by the Hamiltonian of a rigid rotor,

\[ H_R = \hbar B \hat{N}^2. \]  

(3)

\( B \) denotes the rotational constant and \( \hat{N} \) is the orbital angular momentum operator. The eigenstates of \( H_R \) are \( |N,m\rangle \), where \( N = 0,1,2,\ldots \) denotes the angular momentum quantum number, and \( m = -N,-N+1,\ldots,N \) its projection onto the quantization axis. The energies corresponding to the states \( |N,m\rangle \) are \( E_N = \hbar BN (N+1) \). Transitions in the rotational spectrum of diatomic molecules correspond to MW frequencies, and are thus strongly separated from both optical transitions in the electronic level structure, and near-infrared transitions to vibrational excitations of the atomic cores. Due to this separation of energy scales and the resultant decoupling of the corresponding degrees of freedom, in our theoretical description, we can focus on the rotational degrees of freedom alone [47–49]. A key benefit of working with rotational states are long coherence times. Indeed, excited rotational states have lifetimes on the order of a few seconds, which is much longer than relevant experimental time scales.

For diatomic molecules in an external electric field \( \mathbf{e} \), the rigid-rotor Hamiltonian (3) acquires an additional contribution which in the dipole approximation takes the form \(-\mathbf{d} \cdot \mathbf{e}\), where \( \mathbf{d} \) is the dipole moment operator. We thus arrive at the molecular Hamiltonian

\[ H_M = H_R - \mathbf{d} \cdot \mathbf{e} = \hbar B \hat{N}^2 - \mathbf{d} \cdot \mathbf{e}. \]  

(4)

In the following, we choose both the quantization axis and the \( z \)-coordinate axis along the direction of the electric field \( \mathbf{e} = ce_z \), such that the angular momentum projection quantum number \( m \) is conserved. The matrix elements of the molecular Hamiltonian in the basis of eigenstates \( |N,m\rangle \) of the rigid-rotor Hamiltonian Eq. (3) are expressible in terms of Wigner 3-\( j \) symbols as

\[ \langle N, m| H_M |N', m'\rangle = \left[ \hbar BN (N+1) \delta_{N,N'} - D c (-1)^m \sqrt{(2N+1)(2N'+1)} \right. \]

\[ \times \left. \begin{bmatrix} N & 1 & N' \\ 0 & 0 & 0 \end{bmatrix} \right] \delta_{m,m'}, \]  

(5)

where \( D \) is the permanent molecule frame dipole moment. From this representation it is straightforward to numerically determine the eigenstates and eigenergies of the molecular Hamiltonian, which we denote by \( |N,m\rangle_\mathbf{e} \) and \( E_{N,m}(\mathbf{e}) \), respectively. We note that the degeneracy of levels with different angular momentum \( z \)-components \( |m| \) is lifted by the electric field. As illustrated in Fig. 3(a), for \( e \to 0 \), the states \( |N,m\rangle_\mathbf{e} \) and the corresponding energies are adiabatically connected to the eigenstates \( |N\rangle \) of the dipole Hamiltonian Eq. (3), that is, \( |N,m\rangle_\mathbf{e=0} = |N,m\rangle \) and \( E_{N,m}(\mathbf{e=0}) = E_{N,m} \). At finite values of the electric field, the rotational energy levels acquire a Stark shift. The change of the energy \( E_{N,m}(\mathbf{e}) \) with the electric field \( \mathbf{e} \) corresponds to the induced dipole moment

\[ \mathbf{d}^{N,m}(\mathbf{e}) = -\nabla \epsilon \]  

E_{N,m}(\mathbf{e}) = \epsilon |N,m\rangle \mathbf{d}|N,m\rangle_\mathbf{e} \] shown in Fig. 3(b). The induced dipole moment is aligned along the electric field, i.e., along the \( z \) axis.

As indicated above, to engineer spatially varying induced dipole moments, we couple internal and motional degrees of freedom of the molecules by applying electric fields which vary in space. In particular, below we consider an electric field \( \mathbf{e} = e_0 + \mathbf{e}'(\mathbf{r}) \) which is composed of a spatially homogeneous offset field \( \mathbf{e}_0 = e_0 \mathbf{e}_z \) and a gradient field \( \mathbf{e}'(\mathbf{r}) = e'_z \mathbf{e}_z (\mathbf{e}' \cdot \mathbf{r}) \) where \( \mathbf{r} = (x, y, z) \) is the position of the molecule. The gradient field grows linearly in the direction specified by \( \mathbf{e}' \). In particular, we choose \( \mathbf{e}' = \mathbf{e}_x \) and \( \mathbf{e}' = \mathbf{e}_z \) in Secs. IV and V, respectively. In both setups considered below, the motion of the molecules along the direction \( \mathbf{e}' \) is restricted to a region of spatial extent \( a_L \) around the coordinate origin. We assume \( e_0 \gg e' a_L \), so that the gradient field can be taken into account perturbatively. To first order in the gradient field, the Stark shift of the rotational energy levels is

\[ E_{N,m}(\mathbf{e}_0 + \mathbf{e}'(\mathbf{r})) \approx E_{N,m}(\mathbf{e}_0) - |\mathbf{d}^{N,m}(\mathbf{e}_0) \cdot \mathbf{e}'|(\mathbf{e}' \cdot \mathbf{r}). \]  

Higher orders are negligible for large electric field offsets considered in the following sections.
where the matrix elements of $\hat{H}_c$ are typically much smaller than the energy scale $\hbar \omega$ set by the MW frequency. To carry out this simplification, we first eliminate the diagonal elements of $\hat{H}_c$ by the transformation $|\tilde{N},0\rangle_{\epsilon_0} \rightarrow \exp\left[i\omega t\hat{H}_c^{N,N} / \omega\right] |\tilde{N},0\rangle_{\epsilon_0}$. Upon applying this transformation, the off diagonal elements acquire additional phases factors, $H_c^{\tilde{N},N'} \rightarrow H_c^{\tilde{N},N'} \exp\left[i\omega \left(H_c^{\tilde{N},N} - H_c^{\tilde{N},N'}\right) / \omega\right]$, which, however, can be neglected due to the condition $H_c^{\tilde{N},N'} \ll \hbar \omega$. Further, this condition justifies a rotating wave approximation, and in a frame which rotates at the MW frequency $\omega$ and is defined by the transformation

$$U_{2LS}(t) = |0,0\rangle_{\epsilon_0}(0,0) + e^{i\omega t}|2,0\rangle_{\epsilon_0}(2,0)$$

the 2LS Hamiltonian simplifies up to an overall energy shift to

$$H_{2LS}^0 = \hbar \left(\frac{-\Delta_{2,0}}{\Omega/2} \frac{\Omega/2}{\hbar} \right).$$

Here, the Rabi frequency defined as $\Omega/2 = H_{2,0}^{0,2}/\hbar$ is assumed to be real, and the detuning is given by $\Delta_{2,0} = \omega - [E_{2,0}(\epsilon_0) - E_{0,0}(\epsilon_0)] / \hbar + \Delta_{0,0} \ll \Omega$. The detuning includes a component $\Delta_{0,0}$ which can be set zero in the considered case. However, we keep this parameter at this point to anticipate that below we add another static but spatially inhomogenous electric field component. Then, also $\Delta_{0,0}$ becomes a function of position and can be eliminated only at one point in space.

To engineer a potential barrier as described below, we require the extension of the above 2LS to a 3LS which comprises the three rotational states $|0,0\rangle_{\epsilon_0}, |1,0\rangle_{\epsilon_0}$, and $|2,0\rangle_{\epsilon_0}$. As above, we assume the presence of a static background field $\epsilon_0$, but now we add two oscillating components with frequencies $\omega_1$ and $\omega_2$, which couple the
pairs of states $|0,0\rangle_{\epsilon_0}$ and $|\bar{1},0\rangle_{\epsilon_0}$ and $|\bar{1},0\rangle_{\epsilon_0}$ and $|\bar{2},0\rangle_{\epsilon_0}$, respectively. This situation is illustrated in Fig. 3(c). Following the same arguments as in the 2LS, the 3LS Hamiltonian in a rotating frame defined by the transformation

$$U_{3LS}(t) = e^{-i\omega_3 t}|0,0\rangle_{\epsilon_0}|0,0\rangle + |\bar{1},0\rangle_{\epsilon_0}|0,0\rangle + e^{i\omega_3 t}|\bar{2},0\rangle_{\epsilon_0}|\bar{2},0\rangle$$

reads up to an overall energy shift

$$H^0_{3LS} = \hbar \begin{pmatrix} -\Delta_{\bar{2},0} & \Omega_2/2 & 0 \\ \Omega_2/2 & -\Delta_{\bar{1},0} & \Omega_1/2 \\ 0 & \Omega_1/2 & -\Delta_{0,0} \end{pmatrix}. \tag{11}$$

$\Omega_1$ and $\Omega_2$ are the Rabi frequencies associated with the MW driving fields, and the corresponding detunings from the rotational transition frequencies are given by $\Delta_{\bar{2},0} = (\omega_1 + \omega_2) - (E_{\bar{2},0} - E_{0,0})/\hbar + \Delta_{0,0}$ and $\Delta_{\bar{1},0} = \omega_1 - (E_{\bar{1},0} - E_{0,0})/\hbar + \Delta_{0,0}$. As above, the detunings include a component $\Delta_{0,0}$, which we keep as a placeholder for a spatially inhomogenous electric field. From this point on, to simplify the notation, we drop the explicit dependency on $\epsilon_0$ for all quantities.

### IV. INTERFACE BOUND STATE IN EFFECTIVE TWO-LEVEL SYSTEMS

We now consider a setup in which two rotational levels are coupled by a MW field to form an effective two-level system as described by Eq. (9), and additionally a time-independent electric field is applied which is oriented along the $z$ axis and increases linearly in the $x$ direction. As we show, in this setup, bound states of two molecules can form at the interface at which the induced dipole moment changes sign.

#### A. Single-particle physics

The key elements to engineer interface bound states are a position-dependent electric field, $\epsilon(x) = \epsilon_0 + \epsilon' x$, which comprises a homogeneous offset field $\epsilon_0$ and a gradient field $\epsilon' x$, and a MW-induced Rabi coupling of the molecular levels $|0,0\rangle$ and $|\bar{2},0\rangle$ as discussed in Sec. III. As illustrated in Fig. 3(b), the strong offset field $\epsilon_0$ induces state-dependent dipole moments $\mathbf{d}^{N,m}$, which couple to the gradient field $\epsilon' x$ and thus give rise to position-dependent Stark shifts. As a result, the molecules move in a state-dependent linear potential, and the Rabi coupling between the internal states of the molecules is resonant only at a particular point, which we chose as the coordinate origin. The Hamiltonian which describes the motion of a single molecule in this configuration of electric and MW fields is

$$H_{2LS} = \frac{p_x^2}{2m} + H^0_{2LS}(x), \tag{12}$$

where $p_x = -i\hbar \partial_x$ is the $x$-component of the momentum operator $\mathbf{p} = -i\hbar \nabla$. For the moment, we consider only the motion along the $x$ axis and restore the full three-dimensional (3D) form of the Hamiltonian in the next section. $H^0_{2LS}(x)$ is given by the Hamiltonian in Eq. (9) with linearly position-dependent detunings, $\Delta_{N,0}(x) = \Delta_{N,0} x$, where $\Delta_{N,0} = -\mathbf{d}^{N,0} \cdot \epsilon' / \hbar$. Below we find it convenient to parameterize the Hamiltonian as

$$H^0_{2LS}(x) = \hbar \frac{\Omega}{2} \begin{pmatrix} 1 & 1 - \delta & x/s \\ 1 - \delta & 1 & 1 - x/s \\ x/s & 1 - x/s & 1 \end{pmatrix}, \tag{13}$$

where $-\delta = \Delta_{0,0}/\Delta_{2,0} = \epsilon_{2,0}/\epsilon_{0,0}$ is the ratio of induced dipole moments in the states $|0,0\rangle$ and $|\bar{2},0\rangle$, and

$$s = \frac{\Omega}{\Delta_{0,0}(1 + \delta)} \tag{14}$$

corresponds to the length scale which delimits the spatial region in which the Rabi coupling is resonant. Due to its dependence on the position $x$, the Hamiltonian (13) couples internal and external degrees of freedom. Typically, the energy scales associated with the internal degrees of freedom are much larger than those which characterize the motion of molecules. This allows us to treat the internal dynamics in a BO approximation. To wit, we first diagonalize the 2LS Hamiltonian (13); Thereby, we treat the position $x$ as a parameter. The eigenvalues $E_\pm(x)$ of $H^0_{2LS}(x)$ are given by

$$E_\pm(x) = \hbar \frac{\Omega}{2} \left[ 1 - \delta \frac{x}{1 + \delta} \pm \sqrt{1 + (x/s)^2} \right], \tag{15}$$

and the corresponding eigenvectors read

$$|\pm\rangle_x = \sin(\theta_x/2)|\bar{2},0\rangle + \cos(\theta_x/2)|0,0\rangle, \tag{17}$$

$$|\mp\rangle_x = \cos(\theta_x/2)|\bar{2},0\rangle - \sin(\theta_x/2)|0,0\rangle, \tag{16}$$

where $\theta_x = \arctan(s/x)$ and $0 \leq \theta_x \leq \pi$. The states $|\pm\rangle_x$ are superpositions of the rotational states $|0,0\rangle$ and $|\bar{2},0\rangle$ with spatially varying amplitudes. In particular, $|\pm\rangle_x \rightarrow |0,0\rangle$ and $|\pm\rangle_x \rightarrow |\bar{2},0\rangle$ for $x \rightarrow +\infty$ and $x \rightarrow -\infty$, respectively, with the transition occurring in a spatial region of extent $s$. Correspondingly, the limiting behavior of $|\mp\rangle_x$ is given by $|\mp\rangle_x \rightarrow |\bar{2},0\rangle$ and $|\mp\rangle_x \rightarrow |0,0\rangle$ for $x \rightarrow +\infty$ and $x \rightarrow -\infty$, respectively. The BO approximation [50–54] assumes that the internal state of the molecules adiabatically follows the external motion, where the parametric dependence of the internal state on the position is given by Eq. (16). To formalize this idea, we expand the state of a molecule in the eigenbasis (16) of the 2LS Hamiltonian,

$$|\Psi\rangle = \int dx \sum_{\sigma = \pm} \Psi_\sigma(x) |\pm\rangle_x \otimes |\sigma\rangle_x, \tag{17}$$

where $|x\rangle$ is an eigenstate of the position operator. The effective Hamiltonian $H^0_{\text{eff}}$ for the wave functions $\Psi_\sigma(x)$
in the adiabatic basis can be obtained from Eq. (12) by shifting the momentum operator according to $p_z \to p_z - A(x)$, where
\[
A(x) = \frac{i\hbar}{2m} \left( \begin{array}{cc} 1 & -1 \\ 1 & 0 \end{array} \right)
\]
(18)
can be interpreted as a gauge potential,
\[
H_x^{ad, 2LS} = \frac{p_x^2}{2m} - \{p_x, A(x)\} + \frac{A^2(x)}{2m} + \left( \begin{array}{c} E_+(x) \\ 0 \end{array} \right).
\]
(19)
The terms $\{p_x, A(x)\}$ and $A^2(x)$ represent nonadiabatic corrections. Since $A(x)$ is purely off-diagonal, the anticommutator $\{p_x, A(x)\}$ corresponds to a coupling between the + and $-$ BO channels. $A^2(x)$, on the other hand, is diagonal, and gives a contribution to the effective BO potentials $E_{\alpha}(x)$. The Hamiltonian for the motion of a molecule prepared in the + channel is given by
\[
H_x^{ad, 2LS, ++} = \frac{p_x^2}{2m} + E_+(x) + V_{na}(x),
\]
(20)
where the nonadiabatic potential barrier is given by
\[
V_{na}(x) = \frac{A^2_{++}(x)}{2m} = \frac{\hbar^2}{2ms^2} \frac{1}{4 + (x/s)^2}.
\]
(21)

We focus on a parameter regime that is specified below, in which both the channel coupling and the nonadiabatic contribution to the effective BO potential from $A^2(x)$ are negligible. The validity of this approximation is discussed in detail in Appendix C.

The effective BO potential corresponding to the eigenvalue $E_+(x)$ forms a trap. This is because the induced dipole moments $d_0^2$ and $d_0^0$ have opposite sign as can be seen in Fig. 3(b), and thus the position-dependent Stark shifts $\Delta_{0,0}^{d}(x)$ for $x \gtrsim s$ and $\Delta_{2,0}^{d}(x)$ for $x \lapprox -s$ have opposite signs. For $|x| \ll s$ we approximate the BO trapping potential $E_+(x)$ by a harmonic potential,
\[
E_+(x) \approx \hbar \Omega/2 \left[ 1 - x_0^2/s^2 + (x - x_0)^2/s^2 \right] = \Delta E + m\omega_0^2(x - x_0)^2/2.
\]
(22)
The harmonic approximation is characterized by the effective trap frequency $\hbar \omega_0 = \sqrt{\hbar \Omega/2(2ms^2)}$, the position of the minimum $x_0/s = (1 - \delta)/2\sqrt{\delta}$, and the energy shift $\Delta E = \hbar \Omega/2[1 - (x_0/s)^2]$. In this harmonic potential, the size of the ground state is given by $l_0 = \sqrt{\hbar/(m\omega_0)}$, and for self-consistency we require $l_0/s \ll 1$. The effective potential $E_-(x)$ in the BO channel corresponding to the eigenstate $|\rangle_{-} \rangle$ forms an inverted trap, and consequently this channel hosts a continuum of scattering states. The coupling between the BO channels due to the term $\{p_x, A(x)\}$ in Eq. (19) leads to decay of the bound states in the + channel to the continuum in the $-$ channel. The decay rate can be estimated by Fermi’s golden rule as
\[
\frac{\Gamma_{2LS}}{\omega_0} \approx 2\sqrt{\hbar l_0/s} \exp \left[ -8 \left( \frac{s}{l_0} \right)^2 \right].
\]
(23)
A detailed derivation is provided in Appendix C. The adiabatic limit requires $\Gamma_{2LS}/\omega_0 \ll 1$ which is achieved for $l_0/s \ll 1$, in agreement with the harmonic approximation of $E_+(x)$. Because of the exponential factor in the rate Eq. (23) a moderately small ratio of $l_0/s = 1/\sqrt{2}$ is sufficient to obtain a strongly suppressed decay rate of $\Gamma_{2LS}/\omega_0 < 10^{-6}$. In the next section, we focus on the + channel and discuss bound states of pairs of molecules due to spatially inhomogeneous dipole moments.

B. Dipolar interaction and interface bound state

Since the state $|\rangle_{x} \rangle$ defined in Eq. (16) changes from $|\rangle_{x} \rangle \to |0, 0 \rangle$ for $x \to +\infty$ to $|\rangle_{x} \rangle \to |2, 0 \rangle$ for $x \to -\infty$, also the induced dipole moment $d_0^{(j)}(x)$ defined in Eq. (29) below varies in space and takes limiting values with opposite signs given by $d_0^{(j)}(x) = d_0^{(j),0}$ and $d_0^{(j)}(x) \to d_0^{(j),0}$ for $x \to +\infty$ and $x \to -\infty$, respectively. Therefore, two molecules in the + channel experience attractive induced dipolar interactions if they are located on opposite sides of the point $x_0$ where the dipole moment $d_0(x)$ changes sign. As we show in the following, this gives rise to the formation of bound states of two molecules close to $x_0$.

We consider now the full 3D geometry with a harmonic confinement $V(z) = m\omega_0^2 z^2/2$ in the $z$ direction, whereas motion along the $y$ axis is unrestricted. The Hamiltonian for the motion of two molecules $j = 1, 2$ with position coordinates $r_j = (x_j, y_j, z_j)$ and momenta $p_j$ is then given by
\[
H = \sum_{j=1,2} \frac{p_j^2}{2m} + H_{2LS}(x_j) + \frac{1}{2} m \omega_0^2 z_j^2
\]
(24)
\[+ U^{(1)}_{2LS}(t) \otimes U^{(2)}_{2LS}(t) V_{2d}(r_1 - r_2) U^0_{2LS}(t) \otimes U^0_{2LS}(t),
\]
where the dipolar interaction is
\[
V_{2d}(r) = \frac{1}{|r|^3} \left[ d_1 \cdot \hat{r} d_2 \cdot \hat{r} - 3 d_1 \cdot \hat{r} \frac{d_2 \cdot \hat{r}}{|r|^2} \right]
\]
(25)
and the rotating-frame transformation $U^{(j)}_{2LS}(t)$ for the $j$th molecular molecule is defined in Eq. (8). The wave function for two molecules in the + BO channel can be written as
\[
|\Psi \rangle = \int d r_1 d r_2 \Psi(r_1, r_2) |r_1, r_2 \rangle \otimes |+\rangle_{x_1} \otimes |+\rangle_{x_2}.
\]
(26)

In the following, we neglect nonadiabatic corrections to the BO approximation. Upon projecting the Hamiltonian in Eq. (24) to the + channel, the contribution $H_{2LS}^{xx}(x_j)$ is replaced by the effective potential $E_+(x_j)$. 

Further, the dipolar interaction for two molecules in the + channel is given by

\[ V_{dd}^+(r_1, r_2) = x_x(+) \otimes x_x(+) \left[ U_{2LS}^{(1)}(t) \otimes U_{2LS}^{(2)}(t) \right] \times V_{dd}(r_1 - r_2) U_{2LS}^{(j),+}(t) \otimes U_{2LS}^{(j),+}(t) \left| + \right> x_x \otimes \left| + \right> x_x. \]  

(27)

Due to the rotating-frame transformation \( U_{2LS}^{(j)}(t) \), certain contributions to the dipolar interaction acquire rapidly oscillating phase factors. These contributions average to zero, and we only keep the time-independent components. Then, the dipolar interaction can be written as

\[ V_{dd}^+(r_1, r_2) = \frac{d^D_x(x_1)d^D_x(x_2) + 2d^E_x(x_1)d^E_x(x_2)}{|r_1 - r_2|^3} \times \left[ 1 - 3 \left( \frac{z_1 - z_2}{|r_1 - r_2|^2} \right)^2 \right], \]  

(28)

where the “direct” dipole moments are

\[ d^D_x(x) = x_x(+) \left( \hat{2} \cdot 0 \right) \hat{2} \cdot 0 | \bar{2} \cdot 0 \rangle + \left( \hat{0} \cdot 0 \right) \left( \hat{0} \cdot 0 \right) \hat{0} \cdot 0 | \bar{0} \cdot 0 \rangle \right> x_x, \]  

(29)

and the “exchange” moments, which correspond to interaction processes that exchange the internal states of the molecules, read

\[ d^E_x(x) = \left( \hat{2} \cdot 0 \right) \hat{2} \cdot 0 | \bar{2} \cdot 0 \rangle + \left( \hat{0} \cdot 0 \right) \left( \hat{0} \cdot 0 \right) \hat{0} \cdot 0 | \bar{0} \cdot 0 \rangle. \]  

(30)

In the following, we neglect contributions to \( V_{dd}^+(r_1, r_2) \) which involve \( d^E_x(x) \). This is justified since \( | \langle \bar{2} \cdot 0 | \hat{2} \cdot 0 \rangle | \ll |d^D_x|, |d^D_x| \). We note that a corresponding relation does not apply, for example, for the pair of states \( |0 \cdot 0 \rangle \) and \( |1 \cdot 0 \rangle \). We further remark that the diagonal matrix elements of the dipole moment operator which enter Eq. (29) are not affected by the rotating-frame transformation which lead to the 2LS Hamiltonian (9). The position \( x_0 \) of the zero-crossing of \( d^D_x(x) \) coincides with the minimum of \( E_+(x) \) from Eq. (15) given above. For \( l_0 \ll s \) we expand

\[ d^D(x - x_0) \approx -d^D \frac{4^{\delta^2/3} x}{(1 + \delta)^2 s}. \]  

(31)

to linear order in \( x/s \) around \( x_0 \). Here and in the following, the \( x \)-coordinates \( x_{1,2} \) of the molecules are measured from the zero-crossing \( x_0 \) of the induced dipole moment (29). Then, the projection of the two-molecule Hamiltonian (24) to the + channel reads

\[ H_{2LS}^+ = \sum_{j=1,2} \left[ \frac{p_j^2}{2 m} + E_+(x_j) + \frac{1}{2} m \omega^2 \frac{z^2_j}{l_j^2} \right] + V_{dd}^+(r_1, r_2). \]  

(32)

Since the induced dipole moment (29) is oriented along the \( z \) axis, inelastic “head-to-tail” collisions of the two molecules can be suppressed through tight confinement in this direction. We assume that the trapping frequency \( \omega \) is sufficiently large such that excited states in the potential \( V_+(z) \) are energetically inaccessible, and the molecules reside in the ground state. Then, the wave function \( \Psi(r_1, r_2) \) in Eq. (26) factorizes as

\[ \Psi(\rho_1, \rho_2) = \Phi_{ij}(\rho_1, \rho_2) \Phi_1^\dagger(\zeta, \zeta) \Phi_2^\dagger(\zeta, \zeta) \]  

where \( \rho_j = (x_j, y_j) \). The harmonic oscillator ground state wave function reads

\[ \Phi_0(\zeta) = \exp \left( -\frac{\zeta^2}{2\rho_0^2} \right), \]  

(33)

where \( \rho_0^2 = \hbar/(m \omega^2) \) is the corresponding oscillator length. Under these conditions, which also imply \( l_0 \ll l_0 \), the motion of the molecules is confined to the \( xy \) plane, and the system becomes effectively two-dimensional (2D). Up to the zero-point energy in the harmonic confinement in the \( z \) direction, the Hamiltonian for the 2D motion of the two molecules is given by

\[ H_{2D} = \sum_{j=1,2} \left[ \frac{p_j^2}{2 m} + E_+(x_j) \right] + V_{2D}(\rho_1, \rho_2). \]  

(34)

We obtain the effective 2D dipolar interaction by integrating out the tightly confined \( z \) direction, which yields

\[ V_{2D}(\rho_1, \rho_2) = \int dz_1 dz_2 |\Phi_1^0(\zeta_1)|^2 |\Phi_1^0(\zeta_2)|^2 V_{dd}^+(r_1, r_2), \]  

(35)

That is, the effective 2D dipolar interaction can be written as the product of position-dependent dipole moments \( d^D_x(x_j) \) and an interaction potential \( v_{2D}(\rho) \) which depends only on the relative distance in the \( xy \) plane, \( \rho^2 = (x_1 - x_2)^2 + r_y^2 \), where \( r_y = y_1 - y_2 \) is the relative coordinate in the \( y \) direction. The interaction potential is given by

\[ v_{2D}(\rho) = \frac{1}{\sqrt{8 \pi l_0^2}} \exp \left( \frac{\rho^2}{4 l_0^2} \right) \left( 2 + \frac{\rho^2}{l_0^2} \right) K_0 \left( \frac{\rho^2}{4 l_0^2} \right) - \frac{\rho^2}{l_0^2} K_1 \left( \frac{\rho^2}{4 l_0^2} \right). \]  

(36)

Here, \( K_n(z) \) is the modified Bessel function of the second kind. At large distances \( \rho \gg l_0 \), the interaction potential assumes the characteristic dipolar form \( v_{2D}(\rho) \sim 1/\rho^3 \); at short distances \( \rho \ll l_0 \) it diverges logarithmically, \( v_{2D}(\rho) \sim \sqrt{2/\pi} l_0^2 \ln(\rho/l_0) \). With the identification

\[ d_+(x_j) = d^D_x(x_j), \]  

and if we replace the interaction potential by its asymptotic form \( v_{2D}(\rho) \sim 1/\rho^3 \), the Hamiltonian (34) reduces to Eq. (1) in the Introduction.

The setup we consider is translationally invariant along the \( y \) axis and, therefore, the motion of two molecules in this direction factorizes into center-of-mass (COM) and
relative components, \( \Psi_j(\rho_1, \rho_2) = \psi(x_1, x_2, r_y) \xi(R_y) \), where \( R_y = (y_1 + y_2)/2 \) is the COM coordinate. Possible bound states are negative-energy solutions of the two-body Schrödinger equation (SE)

\[
\sum_{j=1,2} \left( \frac{\hbar^2}{2m} \frac{\partial^2}{\partial x_j^2} + \frac{1}{2} m \omega_0^2 x_j^2 \right) - \frac{\hbar^2}{m} \frac{\partial^2}{\partial r_y^2} + V_{2D}(x_1, x_2, r_y) \psi(x_1, x_2, r_y) = (E_B + \hbar \omega_0) \psi(x_1, x_2, r_y), \quad (37)
\]

where we replaced the effective potential \( E_+(x) \) in the + BO channel by its harmonic approximation (22), and we omitted the energy offset \( \Delta E \). The binding energy \( E_B \) is measured from the ground state energy of the noninteracting two particle problem, which is \( 2 \times \hbar \omega_0/2 \). We solve this equation numerically. Details are discussed in Appendix A, and we present our results in Fig. 4. As shown in Fig. 4(a), a bound state occurs for sufficiently strong induced dipolar interactions, where the strength of dipolar interactions is characterized by the ratio \( a_d^{2.0}/l_0 \) with

\[
a_d^{2.0} = m \left[ \frac{4 \delta^{3/2}}{\hbar (1 + \delta)^2} \right]^2. \quad (38)
\]

Figure 4(b) shows the wave function of the bound state for \( l_0 = 3l_1 \) and \( a_d^{2.0} = 35l_0 \). Numerically, we find that the threshold value for the formation of a bound state depends only slightly on \( l_1 \). This is because \( V_{2D}(x_1, x_2, r_y) \) is only modified in regions where \( (x_1 - x_2)^2 + r_y^2 < l_1^2 \), and in these regions, the probability amplitude \( |\psi(x_1, x_2, r_y)|^2 \) is suppressed. We note that the existence of a threshold value of the ratio \( a_d^{2.0}/l_0 \) implies that strong harmonic confinement suppresses the formation of the bound state.

These results can be understood qualitatively within a simplified model which we obtain from Eq. (37) by setting both the COM coordinate in the \( x \) direction, \( R_x = (x_1 + x_2)/2 \), and the relative coordinate in the \( y \) direction to zero, \( R_y = r_y = 0 \). This yields an effective SE for the component of the wave function which describes the relative motion of the two molecules in the \( x \) direction. The corresponding effective potential, which depends only on the relative coordinate \( r_x = x_1 - x_2 \), contains contributions from the harmonic confinement and the dipolar interaction and is given by

\[
V_{\text{eff}}(r_x) = \frac{1}{4} m \omega_0^2 r_x^2 + V_{2D}(r_x/2, -r_x/2, 0). \quad (39)
\]

Figure 4 shows the effective potential for \( a_d^{2.0}/l_0 = 0 \) and \( a_d^{2.0}/l_0 = 35 \). In the former case, \( V_{\text{eff}}(r_x) \) reduces to a harmonic potential. Then, the state with lowest energy is just the corresponding harmonic oscillator ground state. For \( a_d^{2.0}/l_0 = 35 \), the effective potential exhibits two minima at \( r_x = \pm r_{x,0} \neq 0 \). In this situation, it is energetically advantageous for the molecules to “pay the price” of climbing up to the first excited state in the harmonic potential and thus effectively increase their relative distance, since this allows them to reduce their total energy due to the contribution from the dipolar interaction in Eq. (39). Evidently, the formation of a bound state can be suppressed by increasing the strength of the harmonic confinement.

The above simplified model does not take symmetry requirements on the wavefunction for two identical fermions into account. To discuss this point, we return to the full SE (37). We note that a Hamiltonian which describes the motion of identical particles has to be symmetric under the exchange \( \rho_1 \leftrightarrow \rho_2 \) of the coordinates of the particles. The Hamiltonian in Eq. (37) obeys an even stronger symmetry: It is symmetric under the exchange of both only the \( x \) coordinates, \( x_1 \leftrightarrow x_2 \), and only the \( y \) coordinates, \( y_1 \leftrightarrow y_2 \). Therefore, also its eigenfunctions have definite parity under these operations, i.e., \( \psi(x_1, x_2, r_y) = \pm \psi(x_2, x_1, r_y) \) and \( \psi(x_1, x_2, r_y) = \pm \psi(x_1, x_2, -r_y) \). Overall, the two-body wave function for identical fermions has to be antisymmetric, \( \psi(x_1, x_2, r_y) = -\psi(x_2, x_1, -r_y) \). Numerically, we find that the bound state wave function is antisymmetric with respect to \( x_1 \leftrightarrow x_2 \), and symmetric under \( y_1 \leftrightarrow y_2 \) (or, equivalently, \( r_y \rightarrow -r_y \)). Finally, we note that fermionic statistics imply that the probability of a close encounter of the molecules is strongly suppressed, i.e., \( \psi(x_1, x_2, r_y) \rightarrow 0 \) for \( x_1 \rightarrow x_2 \) and \( r_y \rightarrow 0 \). In comparison to bosonic molecules, this enhances the stability of fermionic molecules against chemical reactions. Apart from reduced stability, bound states also occur for pairs of bosonic molecules in the 2LS configuration. However, for bosons we expect an increased threshold value of the dipolar length. This is because to form a bound state, bosons have to populate the second excited harmonic oscillator state for their relative coordinate in the \( x \) direction. The first excited state, which gives the dominant contribution to the bound state of two fermionic molecules, is excluded for bosons by symmetry.

C. Experimental parameters

In this section we discuss the optimal choice of experimental parameters for the realization of an interface bound state. From a single-particle point of view we require the decay rate of molecules in the + channel given in Eq. (23) to be small, \( \Gamma_{2LS}/\omega_0 \ll 1 \), which is guaranteed for \( l_0/s \ll 1 \). However, even a moderately small ratio of \( l_0/s = 1/\sqrt{2} \) leads to \( \Gamma_{2LS}/\omega_0 < 10^{-6} \), such that the lifetime of molecules in the + channel is well above any experimentally relevant time scale. The rela-
fore, the difference between induced dipole moments as large as possible. In current experiments, the elec-

boils down to finding the optimal value of the electric

such that $\Omega$ is fixed for a given value of $l_0$. In comparison, recent experiments with KRb [10] reached temperatures which correspond to a much lower energy of 50 nK $k_B = 2\pi \times 1$ kHz $h$. We list additional parameters for different species of polar molecules in Tab. 1.

We finally point out that the dipolar length scales as $a_d^{2,0}/l_0 \propto D^{2/3} \frac{m^{1/3}}{E_0}$ with a prefactor that depends on the value of the offset field $\epsilon_0$. In experiments, this dependence on $\epsilon_0$ can be used to tune the dipolar length by adjusting the value of $\epsilon_0$, and thus cross the threshold for the formation of a bound state.

### D. Adiabatic loading

To conclude our discussion of the 2LS, we present an experimental protocol to adiabatically prepare molecules in the $+ \rightarrow -$ channel. The electric offset field is kept at a constant value $\epsilon_0$ throughout the protocol, while the gradient field $\epsilon'$ as well as the KRb molecule with parameters $m = 91 u$, $D = 3.99$ Debye. Further, we take the electric field gradient to be $\epsilon' = 5 (kV/cm)/mm$. Then, with the optimal value $\epsilon_0 = 7.5 hB/D$ of the offset field, the harmonic oscillator length evaluates to $l_0 = 32$ mm, which is considerably below optical length scales. For the same parameters, the dipolar length is given by $a_d^{2,0}/l_0 = 78$, which is above the threshold value for the appearance of a bound state. As can be seen in Fig. 4, the corresponding binding energy is on the order of $E_0 = 2\pi \times 44$ kHz $h$ for the given parameters.

We list additional parameters for different species of polar molecules in Tab. 1.

### Table I. A list of relevant parameters for different fermionic polar molecules [56, 57].

| Molecule | $m/u$ | $D$/Debye | $B$/GHz | $\epsilon_0$/(kV/cm) | $a_d^{2,0}/l_0$ | $\Omega$/kHz |
|----------|-------|-----------|---------|----------------------|---------------|-------------|
| KRb [10] | 127   | 0.57      | 1.11    | 28.9                 | 1.3           | 109         |
| NaK [15, 55] | 63 | 2.72      | 2.83    | 15.5                 | 20            | 389         |
| LiRb | 91 | 3.99      | 7.61    | 28.4                 | 78            | 445         |
| LiCs | 139 | 5.39      | 6.54    | 18.1                 | 277           | 472         |

Figure 4. (a) Energy of the bound state in units of $E_0 = \hbar^2/2m a_d^2$ as a function of the dipolar length $a_d^{2,0}$ defined in Eq. (38) for several values of $l_0/l_\perp$ and $s = \sqrt{2} l_0$. (b) Probability density $\int dR_0 |\psi(r_x, R_z, r_y)|^2$ of the bound state, where $r_x = x_1 - x_2$ and $R_z = (x_1 + x_2)/2$ are the relative and COM coordinates in the x direction, for $l_0 = 3l_\perp$ and $a_d/l_0 = 35$. L denotes the box size used in the numerical diagonalization of Eq. (37). (c) Effective potential (39) for the simplified model discussed in the main text.

\[ h\Omega = 2 \left[ \epsilon' a_d^{2,0} (1 + \delta) \frac{\hbar}{\sqrt{m}} \right]^{2/3} \]

such that $\Omega$ is fixed for a given value of $\epsilon'$. This can be used to determine the harmonic oscillator length in the adiabatic regime as

\[ l_0 = \sqrt{\frac{2 \hbar}{m \epsilon'} a_d^{2,0} (1 + \delta)} \left( 1 + \frac{1}{\sqrt{2}} \right)^{1/3} \]

As $l_0$ sets the length and energy scale of the problem, the denominator in the above expression should be as large as possible. In current experiments, the electric field gradient is limited by the apparatus. Therefore, the difference between induced dipole moments $|d_z^{2,0}(1 + \delta)| = |d_z^{2,0} - d_z^{0,0}|$ has to be maximized. This boils down to finding the optimal value of the electric offset field, which is $\epsilon_0 = 7.5 hB/D$. In turn, this implies $\Delta_0^{2,0} = 0.74 D$, $d_z^{2,0} = -0.26 D$ and $\delta = 2.81$. As an example, we consider the LiRb molecule with parameters $m = 91 u$, $D = 3.99$ Debye. Further, we take the electric field gradient to be $\epsilon' = 5 (kV/cm)/mm$. Then, with the optimal value $\epsilon_0 = 7.5 hB/D$ of the offset field, the harmonic oscillator length evaluates to $l_0 = 32$ mm, which is considerably below optical length scales. For the same parameters, the dipolar length is given by $a_d^{2,0}/l_0 = 78$, which is above the threshold value for the appearance of a bound state. As can be seen in Fig. 4, the corresponding binding energy is on the order of $E_0 = 2\pi \times 44$ kHz $h$ for the given parameters. In comparison, recent experiments with KRb [10] reached temperatures which correspond to a much lower energy of 50 nK $k_B = 2\pi \times 1$ kHz $h$. We list additional parameters for different species of polar molecules in Tab. 1.

We finally point out that the dipolar length scales as $a_d^{2,0}/l_0 \propto D^{2/3} \frac{m^{1/3}}{E_0}$ with a prefactor that depends on the value of the offset field $\epsilon_0$. In experiments, this dependence on $\epsilon_0$ can be used to tune the dipolar length by adjusting the value of $\epsilon_0$, and thus cross the threshold for the formation of a bound state.

D. Adiabatic loading

To conclude our discussion of the 2LS, we present an experimental protocol to adiabatically prepare molecules in the $+ \rightarrow -$ channel. The electric offset field is kept at a constant value $\epsilon_0$ throughout the protocol, while the gradient field $\epsilon'$ as well as the 2LS Hamiltonian (9) in the rotating frame are set to zero initially. Further, we assume that the molecules are prepared in the rotational state $|0, 0\rangle$, and that they are trapped in an auxiliary optical potential, which is switched off at the end of the protocol. The initial value of the trapping frequency should be chosen comparably to the effective trapping frequency $\omega_0$ in the $+ \rightarrow -$ channel at the end of the protocol when the gradient field is fully switched on.

The first step of the adiabatic protocol is to turn on the Rabi coupling between the states $|0, 0\rangle$ and $|2, 0\rangle$ in the 2LS Hamiltonian (9). The detunings should be chosen such that $\Delta_\pm = \Delta_0 - \Delta_0 < 0$ and $|\Delta_\pm|/\Omega \gg 1$. In this limit, the state $|0, 0\rangle$ corresponds to the upper eigenstate $|+\rangle$ of the 2LS Hamiltonian. The next step is to adiabatically reduce the magnitude of the detuning to zero.
Here, the requirement for adiabaticity is that the rate of change of the detuning is small as compared to the Rabi frequency, \(|\partial_t \Delta z_0|/\Omega^2 \ll 1\). Under this condition, the molecules remain in the instantaneous excited eigenstate |+\rangle of the 2LS Hamiltonian. At the end of the adiabatic sweep, i.e., for vanishing detuning \(\Delta z_0' = 0\), this state is given by |+\rangle = (\langle 0, 0 | + | 2, 0 \rangle)/\sqrt{2}$. The final step to prepare molecules in the position-dependent + channel state |+\rangle_2 in Eq. (16) is to adiabatically turn off the auxiliary optical potential while ramping up the electric gradient field \(\epsilon'\). This can be done such that the effective harmonic confinement of molecules in the + channel, which arises from the combination of the auxiliary optical potential and the confinement induced by the BO potential in the + channel, stays approximately constant. Then, an upper limit on the rate at which the auxiliary potential is switched off and the gradient field is switched on is set by the frequency \(\omega_0\) in Eq. (22). At the same time, states in the − channel evolve from being trapped by the auxiliary potential to being antitrapped by the inverted BO potential, cf. Fig. 1(c). In this process, due to the coupling terms in the Hamiltonian in Eq. (19), there are narrow avoided crossings between excited motional states in the − channel and states in the + channel, which should be passed diabatically. The gaps in these avoided crossings are determined by matrix elements of channel coupling terms. As shown in Appendix C.2.b, these matrix elements are exponentially small, and the resulting condition of diabaticity can always be met in practice.

V. NANOSCALE POTENTIAL FOR EFFECTIVE THREE-LEVEL SYSTEMS

The interplay of the orientational dependence of dipolar interactions and particular trapping geometries can lead to intriguing many-body physics. A case in point is the double-layer setup with fermionic dipoles considered in Refs. [44, 45]. For dipoles which are oriented in the direction perpendicular to the layers, the interaction between dipoles within a single layer is repulsive, whereas the interaction between dipoles in different layers can be attractive and induce inter-layer pairing. The critical temperature which has to be reached to realize this exotic type of fermionic superfluidity is increased for strong dipolar interactions, which can be achieved by reducing the spacing between the layers. In the following, we describe how fermionic polar molecules can be trapped in a double-layer geometry with sub-optical-wavelength spacing [38–42] by employing electric gradient fields and MW radiation to realize an effective Λ-system as described by Eq. (11).

A. Single-particle physics

A 3D cloud of polar molecules can be split into a stack of 2D layers by applying an optical lattice potential \(V_L(\mathbf{z}) = V_0 \sin^2[\pi k_L (z - z_0)]\), where \(V_0\) determines the depth of the lattice potential and \(k_L\) is the wave vector of the trapping laser. (We assume \(V_0\) to be independent of the rotational state, thus ignoring differences in dynamical polarizabilities [58]. This is legitimate in the situation with a strong electric field gradient as considered here.) A sufficiently strong lattice potential leads to the formation of layers at the positions of the potential minima at \(k_L (z - z_0) = n\pi\) for \(n \in \mathbb{Z}\), which can be adjusted by tuning the phase shift \(k_L z_0\). Below, we choose \(z_0 = 0\). The spacing of the layers is thus determined by the optical wavelength \(\lambda_L/2 = \pi/k_L\). In this section, we show how layers with nanoscale spacing can be realized by splitting a single well of the optical potential.

In the vicinity of the potential minimum at \(z_0\), a harmonic approximation applies, i.e., \(V_L(z) \approx m\omega_z^2 (z - z_0)^2/2\), where \(\omega_z = \sqrt{\langle V_L(\mathbf{z}) \rangle / m}\) with the recoil energy \(E_r = h^2 k_L^2 / (2m)\). The size of the ground state wave function in this potential is \(a_L = 1/(k_L V_0/E_r)^{1/4}\). Our goal is thus to engineer a potential barrier of size \(l \ll a_L\) as illustrated in Fig. 5(a). To this end, we consider the motion of a single molecule along the \(z\) axis, which is described by

\[
H_{3LS} = \frac{p_z^2}{2m} + V_L(z) + H_{3LS}^0(z),
\]

where \(p_z = -i\hbar \partial_z\) denotes the momentum operator. The form of \(H_{3LS}^0(z)\), which incorporates the coupling of the rotational levels \(|0, 0\rangle, |1, 0\rangle,\) and \(|2, 0\rangle\) via MW fields, is given in Eq. (11). The detunings on the diagonal of the 3LS Hamiltonian can be made position dependent by applying a static electric field \(\epsilon(z) = \epsilon_0 + \epsilon'z\) which comprises both a homogeneous offset \(\epsilon_0\) and a gradient field \(\epsilon'z\) as discussed in Sec. III. The offset field induces state-dependent dipole moments \(\mathbf{d} \propto \mathbf{z}\), which in turn couple to the gradient field. This gives rise to position-dependent Stark shifts, and leads in Eq. (11) to the replacement \(\Delta_{N,0} \rightarrow \Delta_{N,0}(z) = \Delta_{N,0}' z\), where

\[
\Delta_{N,0}' = -d \mathbf{N}_0 \cdot \epsilon'/\hbar.
\]

Because different rotational states acquire different Stark shifts, the MW coupling is resonant only around a particular point which we choose as the origin of the coordinate system as illustrated in Fig. 5(b).

We are interested in the regime of strong gradient fields which, via the Stark effect, give rise to steep linear potentials \(\Delta_{N,0}' \mathbf{z}\). Under these conditions, an experimentally feasible optical lattice potential is insufficient to trap the molecules. This problem can be overcome by cancelling the force \(\Delta_{N,0}' \mathbf{z}\) which acts on molecules in the rotational state \(|1, 0\rangle\) by applying an equally strong but opposite force. For example, in polar molecules which carry both an electric and a magnetic dipole moment...
such as RbSr [59], this additional force can be induced by a magnetic gradient field which results in a position dependent Zeeman shift. An alternative route, which we discuss in Appendix B, is Floquet engineering by modulating the Rabi coupling $\Omega$ between the levels $|0,0\rangle$ and $|1,0\rangle$ periodically such that the Floquet Hamiltonian which describes the stroboscopic evolution of the system at multiples of the period of the drive features $\Delta'_1 = 0$. Any of these solutions leads to a simplified effective form of the 3LS Hamiltonian given by
\[
H^{3LS}_{\text{eff}}(z) = \hbar \begin{pmatrix} \Delta' z & \Omega/2 & 0 \\ \Omega/2 & 0 & r\Omega/2 \\ 0 & r\Omega/2 & -r^2\Delta' z \end{pmatrix}. \tag{43}
\]

The values of the parameters $\Delta'$ and $r$ depend on whether an additional external force or Floquet engineering is used to cancel $\Delta'_{1,0}$. For an external force, $\Delta' = \Delta'_{1,0} - \Delta'_{2,0}$ and $r^2 = (\Delta'_{2,0} - \Delta'_{1,0})/\Delta'$, whereas Floquet engineering as discussed in Appendix B leads to $\Delta' = \Delta'_{1,0}$ and $r^2 = (\Delta'_{1,0} + \Delta'_{2,0})/\Delta'$. Moreover, in Eq. (43) we set $\Omega_2 = \Omega$ and $\Omega_1 = r\Omega$, which corresponds to a fine-tuning of the relative strength of the Rabi couplings $\Omega_1$ and $\Omega_2$. This choice of parameters guarantees that the effective $\Lambda$-system supports a zero-energy state. To be specific, the eigenvalues $E_\sigma(z)$ with $\sigma = 0, \pm$ of the Hamiltonian (43) are given by $E_0(z) = 0$ and
\[
E_{\pm}(z) = \frac{\hbar}{2} \left[ \Delta' z \left( 1 - r^2 \right) \pm \sqrt{1 + r^2} \sqrt{\Omega^2 + (\Delta' z)^2 (1 + r^2)} \right]. \tag{44}
\]

The eigenvalues $E_\sigma(z)$ are illustrated in Fig. 5. We consider the limit of slow motion of the molecules, in which the eigenvalues $E_\sigma(z)$ and the eigenstates $|\sigma\rangle_z$ form decoupled BO channels [50–54]. In particular, we focus on the zero-energy BO channel, which can be trapped by an optical potential $V_L(z)$. The corresponding zero-energy eigenstate reads
\[
|0\rangle_z = \sum_{N=0,1,2} c_{N,0}(z) |\bar{N},0\rangle = \frac{1}{\sqrt{1 + r^2}} \frac{1}{\sqrt{1 + (z/l)^2}} \left( -r \sqrt{1 + r^2} (z/l) \right), \tag{45}
\]
where
\[
l = \frac{\Omega \sqrt{1 + r^2}}{2r \Delta'}. \tag{46}
\]

denotes the characteristic length scale on which the structure of $|0\rangle_z$ changes. In particular, for $|z| \gg l$, the zero-energy eigenstate is approximately given by $|0\rangle_z \approx |\bar{1},0\rangle$; the internal states $|0,0\rangle$ and $|2,0\rangle$ give relevant contributions to $|0\rangle_z$ only in a region of size $l$ around the resonant point. In contrast to dark states in atomic $\Lambda$ systems [38, 41, 42], the molecular zero-energy eigenstate $|0\rangle_z$ populates also excited states. However, this does not pose a problem for experimental implementations, since the decay rates of rotational excitations are negligibly small as compared to typical experimental time scales. We note that if the abovementioned Floquet scheme is applied, the effective Hamiltonian (43) should be understood as being written in the interaction picture defined by Eq. (B6), and consequently the interaction-picture transformation $R(t)$ should also be inserted in Eq. (45) such that the zero-energy BO state is given by
\[
|0\rangle_z = \sum_{N=0,1,2} c_{N,0}(z) R(t)|\bar{N},0\rangle.
\]

We proceed to derive the nonadiabatic contributions to the zero-energy BO channel and quantify the coupling to the other channels. To this end, in analogy to Eq. (17)
for the 2LS, we expand the wave function of a molecule in the eigenbasis of the effective Λ-system Hamiltonian,

$$|\Psi| = \int dz \sum_{\sigma=0,\pm} \Psi_\sigma(z)|z\rangle \otimes |\sigma\rangle_z. \quad (47)$$

Consequently, the momentum operator in Eq. (42) transforms as \( p_z \rightarrow p_z - A(z) \) with the gauge potential \( A(z)_{z,\sigma} = -z|\sigma\rangle p_z|\sigma\rangle_z \). The Hamiltonian for the amplitudes \( \Psi_\sigma(z) \) in the adiabatic basis thus reads

$$H_{3LS}^{ad} = \frac{p_z^2}{2m} - \{p_z, A(z)\} + A^2(z)$$

$$+ V_L(z) + \begin{pmatrix} 0 & 0 & 0 \\
0 & E_-(z) & 0 \\
0 & 0 & E_+(z) \end{pmatrix}. \quad (48)$$

The contributions from \( A(z) \) are twofold: Since \( A(z) \) is purely off-diagonal in the adiabatic basis spanned by the states \( |\sigma\rangle_z \), the term \( \{p_z, A(z)\} \) describes nonadiabatic channel couplings; On the other hand, \( A^2(z) \) contains both off-diagonal channel couplings and diagonal contributions which give rise to repulsive potential barriers. Below, we discuss conditions under which the nonadiabatic channel couplings are negligible. Then, the Hamiltonian for the motion of a molecule prepared in the zero-energy channel can be written as

$$H_{3LS,00}^{ad} = \frac{p_z^2}{2m} + V_L(z) + V_{na}(z), \quad (49)$$

where the nonadiabatic potential barrier is given by

$$V_{na}(z) = \frac{A^2_{00}(z)}{2m} = \frac{\hbar^2}{2ml^2} \frac{1}{[1 + (z/l)^2]^2}. \quad (50)$$

The characteristic scale \( l \) from Eq. (46) which defines both the width and the height of the potential barrier \( V_{na}(z) \), is determined by the detuning gradients \( \Delta_{\sigma,0}^L \) and the Rabi frequency \( \Omega \). By choosing these parameters such that \( a_L \ll l \), a single potential well generated by \( V_L(z) \) is split into two sites. This is illustrated in Fig. 6(a), where we show the ground (\( \psi_L(z) \)) and first excited (\( \psi_L(z) \)) state in the zero-energy BO channel, which are located, respectively, to the right and left of the barrier. We obtain the states \( \psi_{L,R}(z) \) by numerically diagonalizing the Hamiltonian in Eq. (49). An analytical discussion of the suppression of wave functions inside the barrier is provided in Appendix C.

The aforementioned Hamiltonian ignores nonadiabatic channel couplings due to the terms \( \{p_z, A(z)\} \) and \( A^2(z) \) in Eq. (48). These couplings induce decay of states in the zero-energy BO channel to the other channels. As illustrated in Fig. 5(c), the linear potentials \( \Delta_{\sigma,0}^L \) give rise to trapping and antitrapping of molecules in the + and − channels, respectively. Therefore, the + channel hosts a discrete set of trapped states, none of which are resonant with the states \( \psi_{L,R}(z) \) in the zero-energy channel if the minimal gap \( \hbar\Omega/2 \) between these channels is larger than the spacing of levels \( \approx \hbar\omega_L \) in the zero-energy channel. In contrast, the antitrapped states in the − channel form a continuum. The decay rate from the zero-energy BO channel to the − channel can be obtained from Fermi’s golden rule. The derivation presented in Appendix C yields

$$\frac{\Gamma_{3LS}}{\omega_L} \approx 2 \left( \frac{l}{a_L} \right)^2 \sqrt{\frac{\omega_L}{\omega}} \exp \left( -2.08 \sqrt{\frac{\omega}{\omega_L}} \frac{l}{a_L} \right). \quad (51)$$

This result is valid in the limit \( l/a_L \gg \omega_L/\omega \), when \( \Gamma_{3LS} \) is exponentially suppressed as illustrated in Fig. 6(b).

In addition to decay of the states \( \psi_{L,R}(z) \) in the zero-energy BO channel into the − channel, which is induced by nonadiabatic corrections to the single-particle Hamiltonian in Eq. (49), the stability of a many-body system that is loaded into the states \( \psi_{L,R}(z) \) is reduced by inelastic collisions which can occur at short distances. This effect is minimized when \( \psi_L(z) \) and \( \psi_R(z) \) are maximally localized on either side of the barrier. As a measure of the degree of localization we calculate the wave function leakage \( \mathcal{O}_{L,R} = \int_0^\infty dz |\psi_{L,R}(z)|^2 \). The leakage is affected by the length scales \( a_L \), \( l \) and \( z_0 \), which determine the shape of the wave functions. For \( z_0/a_L = 0.3 \), in Fig. 6(c) we observe minimal leakage for a range of small values of \( l/a_L \). In particular, leakage is negligible in the limit \( l/a_L \ll 1 \). We obtain a hierarchy of conditions which have to be met by combining the requirements of small leakage and low decay rates (51):

$$1 \gg \frac{l}{a_L} \gg \sqrt{\frac{\omega_L}{\omega}} \quad (52)$$

Engineering barriers to cut a single well into two sites is possible if the above states hierarchy of scales is fulfilled. We emphasize that our scheme has sufficiently many “tuning knobs” to adjust each parameter independently. In the next section, we discuss many-body effects arising from dipolar interactions across the barrier.

**B. Dipolar interaction and two-body bound state**

We now turn to many-body physics of molecules in the 3LS configuration discussed above. As an illustrative example, we consider the motion of two identical fermionic molecules. In particular, we study the formation of bound states between molecules on the left and right of the optical nanoscale barrier. We assume a 3D geometry, in which the motion of molecules is not restricted in the \( xy \) plane. For molecules in the zero-energy BO channel, the position-dependent internal state is given by \( |0_z\rangle \) in Eq. (45). The induced dipole moment of this state is oriented along the static electric field, i.e., along the \( z \) axis. Therefore, the interaction of molecules within one of the layers \( L \) or \( R \) is always repulsive, whereas molecules in different layers experience attractive interactions when
the dipolar interaction which acquire rapidly oscillating by the effective BO potential, $H$ energy channel, the contribution $BO$ channel reads

\[
\psi_L(z) = \int \frac{d^3r}{(2\pi)^3} \Psi(r_1, r_2) |r_1, r_2\rangle \otimes |0\rangle_{z_1} \otimes |0\rangle_{z_2}.
\]

Upon projecting the Hamiltonian in Eq. (53) to the zero-energy channel, the contribution $H_{0,3LS}^0(z_j)$ is replaced by the effective BO potential, $E_0(z_j) + V_{na}(z_j)$, where $E_0(z_j) = 0$ and $V_{na}(z_j)$ is the nanoscale barrier. The explicit form of the single-body contributions to Eq. (53) in the zero-energy BO channel is given in Eq. (49). The dipolar interaction for two molecules in the zero-energy BO channel reads

\[
V^0_{dd}(r_1, r_2) = z_1 |0\rangle \otimes z_2 |0\rangle \left[ U^{(1)}_{3LS}(t) \otimes U^{(2)}_{3LS}(t) \right]
\times V_{dd}(r_1, r_2) U^{(2),\dagger}_{3LS}(t) \otimes U^{(1),\dagger}_{3LS}(t) |0\rangle_{z_1} \otimes |0\rangle_{z_2}.
\]

As above for the 2LS, we neglect the contributions to the dipolar interaction which acquire rapidly oscillating phases due to the rotating-frame transformation $U^{(j)}_{3LS}(t)$. Then, the dipolar interaction can be written as

\[
V^0_{dd}(r_1, r_2) = \frac{d^2(z_1, z_2)}{r_{1,2}^3} \left[ 1 - \frac{3}{r_{1,2}} \left( z_1 - z_2 \right)^2 \right].
\]

$r_{1,2}^3 = (z_1 - z_2)^2 + \rho^2$ denotes the relative distance of the two molecules in 3D, and $d^2(z_1, z_2)$ is the expectation value of the product of dipole moment operators for two molecules in the internal state $|0\rangle_z$ given in Eq. (45). We note that only the $z$-components give nonvanishing contributions. Neglecting oscillating phase factors is equivalent to taking a temporal average. Therefore, $d^2(z_1, z_2)$ can be written as

\[
d^2(z_1, z_2) = z_1 |0\rangle \otimes z_2 |0\rangle \overline{\rho}_{zz} |0\rangle_{z_1} \otimes |0\rangle_{z_2},
\]

with the time-averaged dipole moment operator in the rotating frame,

\[
\overline{\rho}_{zz} = \lim_{T \to \infty} \frac{1}{T} \int_0^T dt \left[ U^{(1)}_{3LS}(t) \otimes U^{(2)}_{3LS}(t) \right] \times \hat{d}_{zz} \hat{d}_{zz} U^{(2),\dagger}_{3LS}(t) \otimes U^{(1),\dagger}_{3LS}(t).
\]

Due to the temporal average, $d^2(z_1, z_2)$ does not factorize into two independent contributions. An explicit expression for $\overline{\rho}_{zz}$ is given in Appendix D. We note that here we assume that the position-dependent Stark shift of the state $|1, 0\rangle$ is cancelled by applying an additional external force and not by the Floquet driving scheme outlined above. For Floquet driving, the zero-energy BO state is modified as discussed below Eq. (45). This, however, affects the time-averaged dipolar interaction only slightly. In summary, the projection of the two-molecule Hamiltonian (53) to the $0$ channel reads

\[
H^0_{2,3LS} = \sum_{j=1,2} \left[ \frac{P_j^2}{2m} + V_L(z_j) + V_{na}(z_j) \right] + V^0_{dd}(r_1, r_2).
\]

We assume in the following that the single-particle energy scales associated with the nanoscale potential are dominant as compared to the dipolar interaction, such that motion of the two molecules in the regime of low energies is restricted to the states $\psi_L(z)$ and $\psi_R(z)$. Under these conditions, the nanoscale potential effectively implements a two-layer geometry, with layers $L$ and $R$ which are parallel to the $xy$ plane. As illustrated in Fig. 5(a), the layer separation is given by the barrier size $l$. Then, the two-body wave function $\Psi(r_1, r_2)$ in Eq. (54) can be written as

\[
\Psi(r_1, r_2) = \sum_{\alpha, \beta = L, R} c_{\alpha \beta} \psi_\alpha(z_1) \psi_\beta(z_2).
\]
Ψ_{\text{rel}}(\rho_1, \rho_2) = \Psi_{\text{rel}}(\rho) \Psi_{\text{COM}}(R) \quad \text{where} \quad \rho = \rho_1 - \rho_2 \quad \text{and} \quad R = (\rho_1 + \rho_2)/2 \quad \text{factors into contributions corresponding to the relative and COM motion. Second, due to the symmetry under rotations around the z axis, the wave function corresponding to the relative motion can be decomposed into radial and angular components,}

\Psi_{\text{rel}}(\rho) = \sum_{m \in \mathbb{Z}} c_{m}(\rho) e^{im\phi}. \quad \text{To find a bound state it is sufficient to consider m_z = 0, since a nonvanishing angular momentum adds an additional repulsive centrifugal barrier, which increases the energy. Then, antisymmetry of the wave function \( \Psi(r_1, r_2) \) in Eq. (54) under the exchange of particles requires that the motional state of the two molecules along the z axis is an antisymmetric superposition of the single-particle states \( \psi_L(z) \) and \( \psi_R(z) \), that is, the coefficients \( c_{\alpha \beta} \) in Eq. (60) are given by \( c_{LL} = c_{RR} = 0 \) and \( c_{LR} = -c_{RL} = 1/\sqrt{2} \), so that}

\[ \Psi(r_1, r_2) = \frac{1}{\sqrt{2}} \chi_0(\rho) \Psi_{\text{COM}}(R) \times [\psi_L(z_1)\psi_R(z_2) - \psi_R(z_1)\psi_L(z_2)]. \quad (61) \]

\[ \text{With this ansatz, the Hamiltonian (53) yields the following SE for the radial component of the two-body wave function:} \]

\[ \left[ -\frac{\hbar^2}{2m} \left( \frac{\partial^2}{\partial \rho^2} + \frac{1}{\rho} \frac{\partial}{\partial \rho} \right) + V_{2D}(\rho) \right] \chi_0(\rho) = E_B \chi_0(\rho). \quad (62) \]

A solution with \( E_B < 0 \) corresponds to a bound state. The effective 2D dipolar interaction is given by

\[ V_{2D}(\rho) = \frac{1}{2} \int d^2z_1 d^2z_2 V_{dd}^0(r_1, r_2) \times [\psi_L(z_1)\psi_R(z_2) - \psi_R(z_1)\psi_L(z_2)]^2. \quad (63) \]

It can be decomposed into “direct” and “exchange” contributions, \( V_{2D}(\rho) = V_{2D}^D(\rho) - V_{2D}^E(\rho) \), which read

\[ V_{2D}^D(\rho) = \int d^2z_1 d^2z_2 \psi_L(z_1)\psi_R(z_2) \times V_{dd}(\rho, z_1, z_2) \psi_R(z_2)\psi_L(z_1), \quad (64) \]

and

\[ V_{2D}^E(\rho) = \int d^2z_1 d^2z_2 \psi_L(z_1)\psi_R(z_2) \times V_{dd}(\rho, z_1, z_2) \psi_L(z_2)\psi_R(z_1). \quad (65) \]

We show the dipolar interaction \( V_{2D}(\rho) \) in Fig. 7(b). In the limit of small spatial wave function overlap which is realized, for example, for \( l/a_L \ll 1 \), the exchange part of the interaction gives a strongly suppressed repulsive contribution. As discussed above, the interaction becomes attractive at short distances, when the dipoles are effectively in a “head-to-tail” configuration.

In Fig. 7(a) and (b), respectively, we show the bound state energy \( E_B \) and the corresponding wave function for different values of the dipolar length \( a_d^{1,0} = m (a_d^{1,0}/\hbar)^2 \). A bound state forms for a sufficiently large value of \( a_d^{1,0} \). In particular, for \( l/a_L = 0.1 \) and \( \epsilon_0 = 7\hbar B/D \), the requirement for the formation of a bound state is \( a_d^{1,0}/a_L \gtrsim 3.5 \).

### C. Experimental parameters

In this section we present experimental parameters for the formation of an intra-layer bound state. From a single particle point of view, we have to fulfill the hierarchy of scales \( 1 \gg l/a_L \gg \sqrt{\omega_L / \Omega} \) from Eq. (52). The first inequality ensures that the nonadiabatic barrier splits a single potential well into two sides; The second inequality guarantees stability against diabatic channel couplings, i.e., \( \Gamma_{3LS}/\omega_L \ll 1 \), see Eq. (51). Our goal is to minimize \( l \) subject to the constraints given by the above hierarchy. In this way, the strength of dipolar interactions between particles on different sides of the barrier is maximized.

In terms of the dimensionless ratios \( l/a \) and \( \omega_L / \Omega \), the Rabi frequency \( \Omega \) and the barrier width \( l \) in Eq. (46) can be expressed as

\[ \hbar \Omega = \frac{l}{a_L} \frac{\Delta aL r}{2r} \]

and

\[ l = \left( \frac{\Omega}{\omega_L a_L} \right)^{2/3} \left( \frac{\sqrt{1+r^2} \hbar^2}{2r \Delta \hbar} \right)^{1/3}, \quad (67) \]

respectively. The barrier width is tunable by the detuning gradient \( \Delta \), which should be as large as possible. If the overall energy tilt is removed by an external force as discussed in Sec. V A, the detuning gradient is given by \( \Delta' = (d_z^2 - d_z^0) \epsilon' \). For a given value of the electric field gradient \( \epsilon' \), the difference between the induced dipole moments is maximized for \( \epsilon_0 \) around \( 7 \hbar B/D \), see Fig. 3(b).

This implies \( \Delta' = 0.43 \mathcal{D} \epsilon' \), \( r^2 = 1.30 \), and \( d_z^0 = 0.16 \mathcal{D} \). As a generic example, we consider a molecule with mass \( m = 100 \text{ u} \) and permanent molecule-frame dipole moment \( \mathcal{D} = 5.5 \text{ Debye} \). Moreover, we choose the field gradient as \( \epsilon' = 10 \text{ (kV/cm)/nm} \), and to fulfill the hierarchy of scales we choose \( l/a_L = 0.1 \) and \( \omega_L/\Omega = 1/300 \), which leads to \( \Gamma_{3LS}/\omega_L < 10^{-2} \). We note that for \( l/a_L > 0.1 \), the exchange terms in Eq. (65) give nonnegligible repulsive contributions to the dipolar interaction. With the above parameters, the barrier width evaluates to \( l = 25 \text{ nm} \), which is considerably below optical length scales. For the same parameters, the dipolar length is given \( a_d^{1,0}/a_L = 4.5 \), and is thus above the threshold value for the appearance of a bound state.

We finally note that the dipolar length scales as \( a_d^{1,0}/a_L \propto \mathcal{D}^2 (\mathcal{D} \epsilon')^{1/3} m^{4/3} \) with a prefactor which depends on \( l/a_L \) and \( \epsilon_0 \). The dependence on \( \epsilon_0 \) gives an experimental handle to tune the value of the dipolar length.
the Rabi frequency $\Omega(\rho)$ from zero to a constant value $\Omega_0/2$. For $\rho \to 0$ the interaction potential diverges logarithmically due to the spatial overlap of $\psi_L(z)$ and $\psi_R(z)$. Parameters are the same as in (a) at $a_d/\lambda_L = 4.5$. Energies and the probability density of $\chi_0(\rho)$ correspond to the left-hand and right-hand axis, respectively, and $L$ denotes the numerical box size. The dashed baseline of the radial probability density indicates the corresponding eigenenergy.

D. Adiabatic loading

In the following, we present an adiabatic protocol to load polar molecules in the zero-energy BO channel. As in the adiabatic loading scheme for the 2LS which is discussed in Sec. IV D, the offset field $\epsilon_0$ is kept constant during the protocol, while the gradient field and all Rabi field components give rise to position dependent Stark shifts, which in turn implies that the MW driving fields have to be ramped up simultaneously; In the latter case, both the gradient field $\epsilon'$ and the external force have to be ramped up simultaneously; In the former case, the gradient field is kept at zero while the periodic driving which is described by the Hamiltonian in Eq. (B3) is turned on adiabatically by increasing the amplitude of the drive $\Omega_d$ from zero to the desired value. Adiabaticity can be retained if the quasienergy of interest is always gapped [60], which is the case if the hierarchy of scales in Eq. (B5) is satisfied. Then, the internal state of a molecule at the end of the adiabatic sweep of $\Omega_d$ is the Floquet eigenstate corresponding to the zero-energy eigenstate of $\overline{H}_{\text{eff}}(z)$ in Eq. (B13). The final step of the protocol is to turn on the electric field gradient $\epsilon'$. In the adiabatic limit, the homogeneous zero-energy eigenstate transforms to the position-dependent eigenvector $|0\rangle_z$ in Eq. (45). As in the adiabatic loading scheme of the 2LS which is discussed in Sec. IV D, when the gradient field is turned on, states in the $-\,$ channel eventually become antitrapped, and the narrow avoided crossings which occur between states in the $-\,$ and zero-energy channels should be traversed diabatically.

VI. CONCLUSIONS

To summarize, in this paper we develop tools to engineer many-body Hamiltonians of dipolar particles such as polar molecules or magnetic atoms where dipole moments and single-particle potentials are modulated spatially on nanoscales. We focus on polar molecules, for which our approach utilizes strong static homogeneous as well as inhomogeneous electric fields, and MW radiation to drive transitions between the long-lived rotational levels of diatomic molecules. The inhomogeneous electric field components give rise to position dependent Stark shifts, which in turn implies that the MW driving fields are resonant with rotational transitions only in regions in space that can be as small as tens of nanometers. Then, due to the separation of scales between the internal rotational transitions and the external motional degrees of freedom of the molecules, we obtain approximately decoupled BO channels in which the internal state of the molecules depends parametrically on their position, and can vary on these small scales. Concomitantly, properties such as the induced dipole moment, which determines the interactions between molecules in a many-body system, vary in space. To illustrate essential features of the resultant many-body models, we study the formation of bound states of pairs of fermionic molecules across interfaces at which the induced dipole moment changes sign, and between molecules on different sides of a nanoscale potential barrier.

We note that our analysis which focuses on polar molecules straightforwardly carries over to magnetic atoms [14, 17–25] if static electric fields and field gradients are replaced by magnetic fields. Both
implementations—with polar molecules in electric fields and magnetic atoms in magnetic fields, respectively—benefit from strong field gradients. For example, the formation of a sharp potential barrier in the 3LS discussed in Sec. V hinges on the hierarchy of scales in Eq. (52), which is satisfied for strong Rabi couplings $\Omega$ and gradient fields $\epsilon'$. We finally comment on prospects to explore many-body physics in the 2LS and 3LS configurations beyond the two-particle bound states we considered in this work. The 3LS naturally realizes a double-layer geometry in the relative distance between particles but also on their dependent dipole moment. More generally, the presence in the 2LS setup, in particular, in view of the spatially calibration temperature of the superfluid transition, and are thus beneficial for the observation of the paired phase. Both in the 2LS and in the 3LS configuration, the occurrence of a bound state for sufficiently strong dipolar interactions gives a handle to control scattering properties. In particular, the two-particle scattering length diverges at the threshold value of the interaction strength for the formation of the bound state. It is an interesting question for future studies to investigate possible many-body phases in the 2LS setup, in particular, in view of the spatially dependent dipole moment. More generally, the presence of an interparticle interaction which depends not only on the relative distance between particles but also on their position in one direction, opens up a new unexploited area in dipolar quantum many-body systems.

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Appendix A: Interface bound state: Details of numerical analysis

The wave function and eigenenergies for the bound state shown in Fig. 4 of the main text are the solutions of a numerical diagonalization of Eq. (37). For all calculations we use the linear approximation of the dipole moment,

$$d^D_x(x) \approx -d^2 \frac{4 \delta^{3/2} x}{(1+\delta)^2 s},$$  \hspace{1cm} (A1)$$

as introduced in the main text. It is convenient to express the SE (37) from the main text also along the $x$ direction in relative and COM coordinates, $\bar{r}_x = x_1 - x_2$ and $R_x = (x_1 + x_2)/2$, respectively. This leads to

$$\frac{\hbar^2}{2ml_0^2} \left[ -\frac{1}{2} \frac{\partial^2}{\partial \bar{r}_x^2} - \frac{\partial^2}{\partial r_y^2} + \left( 2\bar{R}_x^2 + \frac{\bar{r}_y^2}{2} \right) \right] \Psi(\bar{R}_x, \bar{r}_x, \bar{r}_y)$$

$$\begin{aligned}
&= E\Psi(\bar{R}_x, \bar{r}_x, \bar{r}_y), \hspace{1cm} (A2)
\end{aligned}$$

where we measure distances in units of the confinement in the $x$ direction $l_0$, $\bar{r}_x = r_x/l_0$, $\bar{r}_y = r_y/l_0$, and $\bar{R}_x = R_x/l_0$. To obtain the binding energy one has to subtract from $E$ twice the ground state energy of the noninteracting system, i.e., $E_B = E - \hbar \omega_0$. The explicit form of the interaction potential is given by

$$V_{2D}(\bar{R}_x + \frac{\bar{r}_y}{2}, \bar{R}_x - \frac{\bar{r}_y}{2}, \bar{r}_y)$$

$$\begin{aligned}
&= \frac{1}{\sqrt{2\pi}} \frac{a_0 l_0^2}{s^2} \left( \frac{\bar{R}_x^2 - \bar{r}_y^2}{4} \right) \exp \left( -\frac{\bar{r}_y^2 l_0^2}{2 \bar{R}_x^2} \right)
&\times \left[ 2 + \bar{r}_y^2 l_0^2 \right] K_0 \left( \frac{\bar{r}_y^2 l_0^2}{4 \bar{R}_x^2} \right) - \bar{r}_y^2 l_0^2 K_1 \left( \frac{\bar{r}_y^2 l_0^2}{4 \bar{R}_x^2} \right),
\end{aligned}$$

(A3)

where we define $\bar{r}_y = \bar{r}_x + \bar{r}_y^2$. Due to the spatial dependence of the dipole moment $d^D_x(x)$ it is not possible to separate the relative and COM motion along the $x$ axis. It is convenient to expand the COM motion in eigenfunctions of the harmonic potential described by the SE

$$\frac{\hbar^2}{2ml_0^2} \left[ -\frac{1}{2} \frac{\partial^2}{\partial \bar{R}_x^2} + 2\bar{R}_x^2 \right] \Phi_n(\bar{R}_x) = \frac{\hbar^2}{2ml_0^2} 2 \left( n + \frac{1}{2} \right) \Phi_n(\bar{R}_x).$$

(A4)

That is, we use the ansatz

$$\Psi(\bar{R}_x, \bar{r}_x, \bar{r}_y) = \sum_{n=0}^{N} \Phi_n(\bar{R}_x) \phi_n(\bar{r}_x, \bar{r}_y),$$

(A5)

where we only keep the lowest $N$ eigenfunctions in the above harmonic potential. We note that the required cutoff $N$ to reach convergent results for the bound state and bound state energy is usually much smaller than the number of grid points one needs to obtain comparable results from a brute-force discretization in real space. The
above ansatz yields the following SE for the amplitudes $\phi_n(\tilde{r}_x, \tilde{r}_y)$:

$$
\frac{\hbar^2}{2m l_0^2} \sum_{m=0}^{N} \left\{ \left[-2 \frac{\partial^2}{\partial r_x^2} - 2 \frac{\partial^2}{\partial r_y^2} + \frac{1}{2} \varepsilon^2 + 2 \left(n + \frac{1}{2} \right) \right] \delta_{m,n} \right. \\
\left. + \nabla_{2D}^m (\tilde{r}_x, \tilde{r}_y) \right\} \phi_m(\tilde{r}_x, \tilde{r}_y) = (E_B - \hbar \omega_0) \phi_n(\tilde{r}_x, \tilde{r}_y).
$$

(A6)

Amplitudes $\phi_n(\tilde{r}_x, \tilde{r}_y)$ with different $n$ are coupled by the interaction matrix elements

$$
\nabla_{2D}^m (\tilde{r}_x, \tilde{r}_y) = \int_{-\infty}^{\infty} d\tilde{r}_x \Phi_{2D}(\tilde{r}_x) \frac{V_{2D}(\tilde{r}_x + \frac{\tilde{r}_x}{2}, \tilde{r}_x - \frac{\tilde{r}_x}{2}, \tilde{r}_y)}{2 \hbar^2 l_0^2} \Phi_m(\tilde{r}_x) \\
= \frac{1}{\sqrt{2\pi}} \frac{a_0 \varepsilon}{\varepsilon} \frac{r^3}{l_0^4} \exp\left(\frac{\rho^2 r^2}{4 l_0^4} \right) \left[ \left(1 + \frac{\rho^2 l_0^2}{4 l_0^4} \right) K_0 \left(\frac{\rho^2 l_0^2}{4 l_0^4} \right) \right] \\
- \rho^2 l_0^2 K_1 \left(\frac{\rho^2 l_0^2}{4 l_0^4} \right) \frac{1}{4} \sqrt{(n+1)(n+2)} \delta_{m,n+2} \\
+ \left((2n+1) - \frac{\rho^2}{4} \right) \delta_{m,n} + \sqrt{n(n-1)} \delta_{m,n-2}.
$$

(A7)

For the parameters used in Fig. 4, we obtain convergence for $N < 9$, a numerical box size of $|\tilde{r}_{x,y}| < 10$ and a uniform grid of $600 \times 600$ for the two relative coordinates.

### Appendix B: Cancellation of linear potentials through Floquet engineering

As discussed in Sec. VA, an electric gradient field induces a state-dependent potential for the motion of molecules which varies linearly in space. This corresponds to a homogeneous force acting on the molecules, and makes it impossible to trap them optically in the regime of strong gradient fields we are interested in. In this appendix, we introduce a Floquet driving scheme which allows us to effectively cancel the force for a specific internal state. The discussion is split into two parts: In the first part, we introduce the general mechanism to cancel the force in the time-independent effective Floquet Hamiltonian which describes the stroboscopic evolution of the periodically driven system. We obtain the effective Hamiltonian in a high-frequency expansion (HFE), which we apply to the Hamiltonian which describes the internal degrees of freedom of the molecules. In the second part, we show that the HFE is still valid if we take into account the coupling between internal and motional degrees of freedom. To be specific, we show that the rate of resonant transitions between motional states due to the Floquet drive is strongly suppressed.

### 1. Effective Hamiltonian from the high-frequency expansion

The linear potential induced by an electric field $\epsilon(z) = \epsilon_0 + \epsilon' z$ is given by $\Delta \tilde{N}_{01}(\omega) = \Delta \tilde{N}_{01}(\omega) = d_{z0}^{\tilde{z}} \epsilon' z$. For the large values of the gradient field $\epsilon'$ we are interested in, the force associated with the linear potential makes it impossible to trap molecules in a far off-resonant optical trap. To overcome this problem, we introduce a Floquet scheme to cancel the induced dipole moment on average. We note that one could also choose the offset field $\epsilon_0$ such that $d_{z0}^{\tilde{z}} = 0$, but this would drastically reduce the strength of dipolar interactions across the barrier. In contrast, through Floquet engineering we achieve that the average dipole moment which determines the force acting molecules is zero, while the dipolar interaction, which is proportional to the average of the square of the dipole moment, is nonzero.

Our starting point is the Hamiltonian in Eq. (11), where we supplement the Rabi frequency $\Omega_1$ for the transition between the rotational levels $|0, 0\rangle$ and $|1, 0\rangle$ with a component with amplitude $\Omega_{1t}^d$ that is periodically modulated with a frequency $\omega_{dt}$, i.e., we substitute $\Omega_1 \rightarrow \Omega_{1t} + \Omega_{1t}^d \cos(\omega_{dt} t)$. In Floquet theory, the evolution of such a periodically driven system from an initial state $|\psi(t_i)\rangle$ at time $t_i$ to a final state $|\psi(t_f)\rangle$ at $t_f$ is written as $|\psi(t_f)\rangle = U(t_f \rightarrow t_i)|\psi(t_i)\rangle = e^{iK(t_f) - iH_{\text{eff}}(t_f - t_i)} e^{iK(t_i)} |\psi(t_i)\rangle$, where $H_{\text{eff}}$ is an effective time-independent Hamiltonian which governs the long-time dynamics of the system, whereas the “kick operator” $K(t)$, which is periodic with period $T = 2\pi/\Omega_{1t}$, describes micromotion within a single driving period. The main idea is now to use the fact that the dipole moments $d_{z0}^{\tilde{z}}$ and $d_{z0}^{\tild{z}}$ of the states $|0, 0\rangle$ and $|1, 0\rangle$, respectively, have different sign, which allows us to mix them in such a way that the effective dipole moment which occurs in $H_{\text{eff}}$ vanishes. In this way, we recover the effective Hamiltonian in Eq. (43). To derive an explicit expression for the effective Hamiltonian, we employ the HFE as discussed in Refs. [61, 62]. We first consider only the internal degrees of freedom and treat the spatial coordinate $z$ as a parameter. In the next section, we discuss corrections to the HFE due to the motional degrees of freedom. The time-dependent Hamiltonian thus reads

$$
H_{\text{3LS}}(z, t) = H_{\text{3LS}}^0(z) + H_d \cos(\omega_{dt}),
$$

(B2)

where

$$
H_{\text{3LS}}^0(z) = \begin{pmatrix}
-d_{z0}^{\tilde{z}} & \Omega_d/2 & 0 \\
\Omega_d/2 & -(d_{z0}^{\tilde{z}} + d_{z0}^{\tild{z}}) & \Omega_d/2 \\
0 & \Omega_d/2 & -(d_{z0}^{\tild{z}} - d_{z0}^{\tild{z}})
\end{pmatrix}
$$

(B3)
dipole moments, \(d\), assuming the hierarchy of scales frequency, and therefore all further calculations are done with the energy scales of the problem being the driving amplitude and the matrices \(\sigma\) and the Pauli matrix \(\tilde{x}\), which are proportional to, respectively, the identity matrix and the Pauli matrix \(\sigma_z\) in the lower block of the Hamiltonian. We assume that the dominant terms of the Hamiltonian is to transform into an interaction picture, in energy scales. The first step to calculate the effective Hamiltonian is to transform into an interaction picture with respect to \(H_0{\text{cos(\omega dt)}}\), defined by the unitary \(R(t) = \exp[\text{−}i\sin(\omega dt)H_d/\omega d]\). The SE in the interaction picture reads

\[
\frac{i\hbar}{\omega d} \partial_t |\psi(t)\rangle = R(t) H_{3LS}(z) R(t) |\psi(t)\rangle = H(t, z, t) |\psi(t)\rangle,
\]

where the interaction-picture Hamiltonian \(H(t, z, t)\) is given by

\[
H(t, z) = \sum_{n \neq 0} V_{I}^{(n)}(z) e^{in\omega dt},
\]

\[
V_{I}^{(n)}(z) = \begin{cases} 
-d_{z}^{0} e^{z} J_n(\frac{\Omega d}{2}) \sigma_{z, 3} + \Omega d/2 J_n(\frac{\Omega d}{2}) \sigma_{x, 1} & \text{for } n \text{ even,} \\
-i d_{z}^{0} e^{z} J_n(\frac{\Omega d}{2}) \sigma_{y, 3} - i \frac{\Omega d}{2} J_n(\frac{\Omega d}{2}) \sigma_{y, 2} & \text{for } n \text{ odd},
\end{cases}
\]

with the time-independent component

\[
\Pi_I(z) = \begin{pmatrix} 
-d_{z}^{0} e^{z} & \Omega d/2 J_0(\Omega d/2) & 0 \\
\Omega d/2 J_0(\Omega d/2) & -d_{z}^{0} e^{z} & \Omega d/2 \\
0 & \Omega d/2 & -d_{z}^{0} e^{z} J_0(\Omega d/2)
\end{pmatrix},
\]

and the matrices

\[
\sigma_{x, 1} = \begin{pmatrix} 0 & 1 & 0 \\
1 & 0 & 0 \\
0 & 0 & 0
\end{pmatrix}, \quad \sigma_{y, 2} = \begin{pmatrix} 0 & 0 & -i \\
0 & 0 & i \\
-i & i & 0
\end{pmatrix}, \quad \sigma_{y, 3} = \begin{pmatrix} 0 & 0 & 0 \\
0 & 0 & -i \\
i & i & 0
\end{pmatrix} \text{ and } \sigma_{z, 3} = \begin{pmatrix} 0 & 0 & 0 \\
0 & 1 & 0 \\
0 & 0 & -1
\end{pmatrix}.
\]

Finally, in terms of the Fourier coefficients \(V_{I}^{(n)}(z)\), the effective Hamiltonian can be written as [61]

\[
H_{\text{eff}}(z) = \Pi_I(z) + \frac{1}{\omega d} \sum_{n=1}^{\infty} \frac{1}{n} \left[V_{I}^{(n)}(z), V_{I}^{(-n)}(z)\right] + O(\|V_{I}^{(n)}\|^3/\omega d^2).
\]
Since the Fourier coefficients \((B9)\) obey \(V_I^{(n)}(z) = V_I^{(-n)}(z)\), the commutator in the lowest-order correction vanishes, and higher-order corrections are at least of order \(O\left(\|V_I^{(n)}\|^3/\omega_d^2\right)\). The matrix norm of the Fourier coefficients \((B9)\) is determined by the parameters \(d_1^2\epsilon_z^2\) and \(\Omega_d\). Therefore, the hierarchy in Eq. \((B5)\) ensures that higher-order corrections to the HFE are suppressed, and we restrict ourselves to the lowest order which is given by the time average of \(H_I(z, t)\), i.e., \(\overline{H}_{I}(z)\). Our goal is thus to find drive parameters \(\Omega_d^2\) and \(\omega_d\) such that the effective dipole moment of the state \(|\overline{I}, 0\rangle\) in the rotating frame defined by Eq. \((B6)\) vanishes,

\[
d_1^2 + d_2^2 J_0 \left(\Omega_d^2/\omega_d\right) = 0.
\] (B12)

A value of the \(\omega_d/\Omega_d^2\) which satisfies this condition can always be found if \(0 \leq -d_1^2/d_2^2 \leq 1\). Then, the effective Hamiltonian becomes

\[
\overline{H}_{\text{eff}}(z) = \begin{pmatrix}
-d_2^2 J_0\epsilon_z^2 & \Omega_2/2 J_0(\Omega_d^2/2\omega_d) & 0 \\
\Omega_2/2 J_0(\Omega_d^2/2\omega_d) & 0 & \Omega_1/2 \\
0 & \Omega_1/2 & -2d_1^2\epsilon_z^2
\end{pmatrix},
\] (B13)

which is equivalent to Eq. \((43)\) in Sec. \(V\ A\) with the identifications \(\Delta' = d_2^2 J_0\epsilon_z^2\), \(\Omega = \Omega_2 J_0(\Omega_d^2/2\omega_d)\), \(r\Omega = \Omega_1\) and \(\text{Im}\Delta' = 2d_1^2\epsilon_z^2\). As desired, the average dipole moment of \(\overline{H}_{\text{eff}}(0)|\overline{I}, 0\rangle\) is removed on a region around the origin which contains the effective system size \(a_L\). Consequently, molecules in this state can be trapped with a far-off-resonant optical trap.

2. Resonant coupling of motional states

In the derivation of the effective Hamiltonian described above, we omitted the motional degrees of freedom of the molecules. To be specific, the contribution to the Hamiltonian which comes from the kinetic energy \(K = -\hbar^2\partial^2/2m\) does not commute with the Fourier coefficients \(V_I^{(m)}(z)\) in Eq. \((B9)\) because of their spatial dependence. This leads to additional corrections to the HFE \((B11)\). To quantify the effect of these corrections, we rewrite the 3LS Hamiltonian \((42)\) with a periodically modulated Rabi frequency \(\Omega_2\) in the interaction picture defined by Eq. \((B6)\) as

\[
H_{3LS}(t) = \frac{\hbar^2}{2m} + V_L(z) + H_{\text{eff}}(z) + V(z, t)
\] (B14)

where

\[
V(z, t) = \sum_{n \neq 0} V_I^{(n)}(z) e^{i\pi\omega_d t},
\] (B15)

and we treat \(V(z, t)\) as a perturbation which induces decay of the unperturbed states with a rate that can be estimated by Fermi’s golden rule. We are interested in the unperturbed states of lowest energy in the zero-energy BO channel, which can be written as \(|L, R\rangle = \int dz \psi_{L,R}(z)|z\rangle \otimes |0\rangle_z\), where the internal state is given below Eq. \((45)\). However, to obtain an estimate of the decay rate, it is sufficient to ignore the nonadiabatic potential in Eq. \((49)\) and consider the ground state wave function of \(K + V_L(z)\), which is the harmonic oscillator wave function

\[
|\phi_0\rangle = \int dz \phi_0(z)|z\rangle \otimes |0\rangle_z
\] (B16)

with

\[
\phi_0(z) = \frac{1}{\sqrt{a_L\sqrt{\pi}}} e^{-z^2/2a_L^2}.
\] (B17)

This state is coupled resonantly to a highly excited scattering state in the 0 channel, which we approximate by a plane wave in a box of size \(L\),

\[
|k\rangle = \int dz \frac{e^{ikz}}{\sqrt{L}}|z\rangle \otimes |0\rangle_z.
\] (B18)

We note that the box size \(L\) drops out of the final result given in Eq. \((B23)\) below. Fermi’s golden rule applies for resonant coupling between the states \(|\phi_0\rangle\) and \(|k\rangle\). The resonance condition for the \(n\)th Fourier component \(V_I^{(n)}(z)\) in Eq. \((B15)\) reads \(\hbar^2 k^2/2m + \hbar\omega_L/2 - n\hbar\omega_d \approx \hbar^2 k^2/2m - n\hbar\omega_d = 0\). For a given harmonic \(n\), this condition determines the value of the wave number \(k = \sqrt{2mn\omega_d/\hbar}\). The corresponding decay rate reads

\[
\Gamma^{(n)}_{k/\omega_L} = \frac{2\pi}{\hbar\omega_L} \left|\langle \phi_0|V_I^{(n)}(z)|k\rangle\right|^2 g(E = n\hbar\omega_d),
\] (B19)

where the density of states in a 1D box of length \(L\) is given by

\[
g(E) = \frac{L}{\hbar\pi} \sqrt{\frac{m}{2E}}.
\] (B20)

Because \(\int dz (0)V_I^{(1)}(z)|0\rangle_z = 0\), the lowest order coupling matrix element is within the hierarchy of scales Eq. \((52)\).
given by

\[
\left|\langle \phi_0 | V_{1L}^{(2)}(z) | 0 \rangle \right|^2 = \int_{-\infty}^{\infty} dz \, \phi_0(z) \langle z | V_{1L}^{(2)}(z) | 0 \rangle \, \frac{e^{ikz}}{\sqrt{L}}^2 
\]

\[
\approx \frac{1}{L} \int_{-\infty}^{\infty} dz \, \phi_0(z) \langle z | V_{1L}^{(2)}(z) | 0 \rangle \, \frac{e^{ikz}}{\sqrt{L}}^2 
\]

\[
+ \Omega_1 \langle z | V_{1L}^{(2)}(z) | 0 \rangle \, \frac{\pi r}{\sqrt{1 + r^2}} e^{-2kl},
\]

where \( l \) denotes the width of the nanoscale barrier from the Eq. (46) and the matrix element is given by

\[
\langle z | V_{1L}^{(2)}(z) | 0 \rangle = d_1^2 e^{-z} z J_2 \left( \frac{(1 + r^2)(z/l)^2 - 1}{(1 + r^2)[1 + (z/l)^2]} \right)
\]

\[
+ \Omega \langle z | V_{1L}^{(2)}(z) | 0 \rangle \, \frac{\pi r}{\sqrt{1 + r^2}} e^{-2kl},
\]

Therefore, the decay rate

\[
\Gamma_{k}^{(2)} = \frac{2\pi}{\hbar \omega_L} \frac{l}{a_L} \frac{1}{\sqrt{\pi}} \left[ d_1^2 e^{-z} z J_2 \left( \frac{(1 + r^2)(z/l)^2 - 1}{(1 + r^2)[1 + (z/l)^2]} \right) \right]
\]

\[
+ \Omega \langle z | V_{1L}^{(2)}(z) | 0 \rangle \, \frac{\pi r}{\sqrt{1 + r^2}} e^{-2kl},
\]

is exponentially suppressed if \( 2kl \gg 1 \) in agreement with Eq. (B5).

**Appendix C: Validity of the Born-Oppenheimer approximation**

Due to the large separation of typical energy scales between the internal rotational and the external motional degrees of freedom of polar molecules, the internal state of a molecule follows its motion essentially adiabatically. In particular, the internal state changes according to the spatial variation of applied electric fields. Deviations from such fully adiabatic dynamics in decoupled BO channels are of two different types: (i) There are nonadiabatic corrections which are diagonal in the basis of BO channels. These corrections are contained in the terms \( A^2(z)/(2m) \) and \( A^2(z)/(2m) \) in Eqs. (19) and (48) for the 2LS and the 3LS, respectively. In our treatment of the 2LS, we assumed that these corrections are negligible, while they give rise to a potentially rather strong nanoscale potential barrier in the 3LS. (ii) Moreover, in both systems, nonadiabatic channel couplings, correspond to the terms \( C_{na} = -\{p_x, A(x) \}/(2m) \) and \( \{p_x, A(z) \} + A^2(z) \) in Eqs. (19) and (48), respectively. In this appendix, we confirm our assumptions concerning nonadiabatic corrections. First, we discuss the impact of diagonal nonadiabatic corrections on wave functions in the BO channels of interest, which are the channel in the 2LS and the zero-energy channel in the 3LS. Second, we study the decay of states in these BO channels due to nonadiabatic channel couplings. We estimate the corresponding decay rates using Fermi’s golden rule, and show that they are exponentially suppressed.

**1. Diagonal nonadiabatic corrections**

The dynamics of molecules in the + BO channel in the 2LS and in the zero-energy channel in the 3LS is described by the Hamiltonians in Eqs. (20) and (49), respectively. Both Hamiltonians take the same form if we treat the effective BO potential \( E_{+}(z) \) in the + channel in the harmonic approximation (22) and neglect the energy shift \( \Delta E \). Then, the generic Hamiltonian in the BO channel of interest reads

\[
H = -\frac{\hbar^2}{2m} \frac{d^2}{dz^2} + 2\frac{m\omega_L^2}{z^2} + \frac{\hbar^2}{2m} \left( 1 - \lambda^2 \right),
\]

where the last term corresponds to the diagonal nonadiabatic correction. The parameter \( \lambda = \sqrt{3}/2 \), and one should replace the \( z \) coordinate by \( x \), \( \omega_L \) by \( \omega_0 \), and \( l \) by \( s \).

In the 2LS, we are interested in the limit \( l_0/s \ll 1 \), where \( l_0 = \sqrt{\hbar/(m\omega_0)} \) is the characteristic length scale associated with the frequency \( \omega_0 \). This inequality implies that the spacing of energy levels in the harmonic potential in Eq. (C1) is much larger than the strength of the nonadiabatic potential, \( \hbar\omega_0 \gg \hbar^2/(2ms^2) \). Under this condition, the nonadiabatic potential is indeed only a negligibly small perturbation.

In contrast, in the 3LS, we require the inverted relation \( a_L \gg l \), where \( a_L \) is the length scale associated with \( \omega_L \). This condition guarantees that the nonadiabatic potential barrier is much narrower than a single well in an optical lattice potential as illustrated in Fig. 5(a).

As we show in the following, this condition also implies that low-energy eigenstates of the Hamiltonian (C1) are strongly suppressed inside the barrier. We thus consider the eigenvalue equation of the Hamiltonian (C1),

\[
H \psi_\lambda(z) = E \psi_\lambda(z).
\]

It is convenient to introduce the new independent variable \( \tau = z/l \) and rewrite this equation as

\[
\left[ \frac{d^2}{d\tau^2} + \frac{t^4}{a_L^4} \tau^2 + \frac{1 - \lambda^2}{(1 + \tau^2)^2} \right] \psi_\lambda(\tau) = \frac{2mL^2}{\hbar^2} E \psi_\lambda(\tau).
\]

For energies smaller than the height of the barrier, \( E \ll \hbar^2/(2ms^2) \), and inside the barrier, \( |\tau| \leq 1 \), one can neglect both the harmonic potential and the right-hand side of this equation, such that the behavior of low-energy wave functions becomes energy independent. To establish this universal behavior, we consider the zero-energy solution of the SE (C3),

\[
\left[ \frac{d^2}{d\tau^2} - \frac{1 - \lambda^2}{(1 + \tau^2)^2} \right] \varphi_\lambda(\tau) = 0.
\]
We can find a general solution of this equation by introducing the new independent variable $y = \arctan(\bar{z})$, such that the equation takes the form
\[
\left[ \frac{d^2}{dy^2} - 2 \tan(y) \frac{d}{dy} - (1 - \lambda^2) \right] \varphi_\lambda(\bar{z}) = 0. \tag{C5}
\]
After introducing the new unknown function $g_\lambda(y) = \varphi_\lambda(y)/\cos(y)$, we finally obtain
\[
\left( \frac{d^2}{dy^2} - \lambda^2 \right) g_\lambda(y) = 0. \tag{C6}
\]
This equation can easily be solved and the general solution of Eq. (C4) then reads
\[
\varphi_\lambda(\bar{z}) = \sqrt{1 + \bar{z}^2} \left\{ A \cos[\lambda \arctan(\bar{z})] + \frac{B}{\lambda} \sin[\lambda \arctan(\bar{z})] \right\} \tag{C7}
\]
where $A$ and $B$ are unknown constants. The general solution $\varphi_\lambda(\bar{z})$ can be used to formulate the effective “boundary conditions” which connect the wave function and its derivative on different sites of the barrier. These conditions are applicable for wave functions which change on a scale that is much larger than the width of the barrier. In other words, the corresponding eigenenergies are much smaller than the height of the barrier. The “boundary conditions” are given by
\[
\begin{align*}
\psi_\lambda'(0^+) + \psi_\lambda'(0^-) &= s\lambda \tan\left(\frac{\pi\lambda}{2}\right) [\psi_\lambda'(0^+) - \psi_\lambda'(0^-)] \\
\psi_\lambda(0^+) - \psi_\lambda(0^-) &= -s\lambda \cot\left(\frac{\pi\lambda}{2}\right) [\psi_\lambda'(0^+) + \psi_\lambda'(0^-)],
\end{align*}
\tag{C8}
\]
where $\psi_\lambda(0^\pm)$ and $\psi_\lambda'(0^\pm)$ are the values of the wave function and its derivative, respectively, on the right and left side of the barrier.

To derive the above conditions, let us consider the asymptotics of $\varphi_\lambda(\bar{z})$ for $\bar{z} \gg 1$,
\[
\varphi_\lambda(\bar{z}) \approx \left[ A\lambda \sin\left(\frac{\pi\lambda}{2}\right) - B \cos\left(\frac{\pi\lambda}{2}\right) \right] \bar{z} + \left[ A \cos\left(\frac{\pi\lambda}{2}\right) + \frac{B}{\lambda} \sin\left(\frac{\pi\lambda}{2}\right) \right] \bar{z}, \tag{C9}
\]
and for $\bar{z} \ll -1$
\[
\varphi_\lambda(\bar{z}) \approx \left[ A\lambda \sin\left(\frac{\pi\lambda}{2}\right) + B \cos\left(\frac{\pi\lambda}{2}\right) \right] \bar{z} + \left[ -A \cos\left(\frac{\pi\lambda}{2}\right) + \frac{B}{\lambda} \sin\left(\frac{\pi\lambda}{2}\right) \right] \bar{z}. \tag{C10}
\]
These asymptotics have to be matched with the wave function and its derivatives for $\bar{z} \sim \pm 1$, $\psi(\bar{z}) \approx \psi_\lambda(0^\pm) + \psi_\lambda'(0^\pm)$. This gives
\[
\begin{align*}
\psi_\lambda(0^+) &= A\lambda \sin\left(\frac{\pi\lambda}{2}\right) + B \cos\left(\frac{\pi\lambda}{2}\right), \\
\psi_\lambda'(0^+) &= \pm A \cos\left(\frac{\pi\lambda}{2}\right) + \frac{B}{\lambda} \sin\left(\frac{\pi\lambda}{2}\right). \tag{C11}
\end{align*}
\]
After excluding the unknown constants $A$ and $B$, and restoring the original units, we arrive at the conditions presented in Eq. (C8).

The boundary conditions in Eq. (C8) allow us to estimate the suppression of the wave function in the barrier region. For this purpose we restore the harmonic potential in Eq. (C1) characterized by the frequency $\omega_L$ and the harmonic length $a_L$, which we assume to be much larger than the width of the barrier $l$, $a_L \gg l$. The eigenenergies and eigenfunctions of this problem can be obtained by matching the solution
\[
y_+ (\bar{z}) = C_+ \exp(-\bar{z}^2/2) \left[ \frac{1}{\Gamma(\frac{1-v}{2})} \Phi\left(\frac{\nu}{2};\frac{1}{2};\bar{z}^2\right) - \frac{2\bar{z}}{\Gamma(-\frac{v}{2})} \Phi\left(\frac{1-v}{2};\frac{3}{2};\bar{z}^2\right) \right] \tag{C12}
\]
which decays exponentially for $\bar{z} \to +\infty$ where $\bar{z} = z/a_L$, with the solution
\[
y_- (\bar{z}) = C_- \exp(-\bar{z}^2/2) \left[ \frac{1}{\Gamma(\frac{1-v}{2})} \Phi\left(\frac{\nu}{2};\frac{1}{2};\bar{z}^2\right) + \frac{2\bar{z}}{\Gamma(-\frac{v}{2})} \Phi\left(\frac{1-v}{2};\frac{3}{2};\bar{z}^2\right) \right] \tag{C13}
\]
which decays exponentially for $\bar{z} \to -\infty$, in the region of the barrier. Here, $\Gamma(\bar{z})$ is the Gamma function, $\Phi(a; b; \bar{z})$ the degenerate hypergeometric function, and $\nu = 2E/\hbar\omega_L$, the energy measured in units of the harmonic oscillator spacing. For low-energy eigenstates, $E \ll \hbar^2/(2mL^2)$, we can use the boundary conditions (C8), which give the following equations:
\[
\begin{align*}
&\left[ \frac{1}{\Gamma(\frac{1-v}{2})} + \frac{1}{a_L \Gamma(-\frac{v}{2})} \lambda \tan\left(\frac{\pi\lambda}{2}\right) \right] (C_+ + C_-) = 0, \tag{C14}
\end{align*}
\]
\[
\begin{align*}
&\left[ \frac{1}{\Gamma(\frac{1-v}{2})} \lambda \tan\left(\frac{\pi\lambda}{2}\right) - \frac{1}{a_L \Gamma(-\frac{v}{2})} \right] (C_+ - C_-) = 0. \tag{C15}
\end{align*}
\]
These equations determine the eigenenergies $\nu$. The eigenenergies of states which are symmetric with respect to $\bar{z} \to -\bar{z}$ fulfill
\[
\frac{1}{\Gamma(\frac{1-v}{2})} + \frac{1}{a_L \Gamma(-\frac{v}{2})} \lambda \tan\left(\frac{\pi\lambda}{2}\right) = 0, \tag{C16}
\]
where $C_+ = C_-$ follows from Eq. (C14). The correspond-
ing eigenfunctions for $|z| \geq l$ have the form
\begin{equation}
\begin{split}
y_{\text{sym}}(z) & \sim \exp\left(\frac{-z^2}{2a_L^2}\right) \left[ \frac{1}{a_L} \lambda \tan\left( \frac{\pi \lambda}{2} \right) \right. \\
& \times \Phi\left(-\frac{\nu}{2}, \frac{z^2}{a_L^2}\right) + \left. \frac{z}{a_L} \Phi\left(1 - \frac{\nu}{2}, \frac{3z^2}{2a_L^2}\right) \right].
\end{split}
\end{equation}
(C17)

The eigenenergies of antisymmetric solutions, for which $C_+ = -C_-$ results from Eq. (15), satisfy the equation
\begin{equation}
\begin{split}
\frac{1}{\Gamma\left(-\nu, \frac{\pi \lambda}{2}\right)} \lambda \tan\left( \frac{\pi \lambda}{2} \right) - \frac{l}{a_L} \frac{2}{\Gamma\left(-\frac{\pi \lambda}{2}\right)} = 0,
\end{split}
\end{equation}
and the wave functions for $|z| \geq l$ are
\begin{equation}
\begin{split}
y_{\text{asym}}(z) & \sim \exp\left(\frac{-z^2}{2a_L^2}\right) \left[ \text{sign}(z) \frac{1}{a_L} \lambda \tan\left( \frac{\pi \lambda}{2} \right) \right. \\
& \times \Phi\left(-\frac{\nu}{2}, \frac{z^2}{a_L^2}\right) + \left. \frac{z}{a_L} \Phi\left(1 - \frac{\nu}{2}, \frac{3z^2}{2a_L^2}\right) \right].
\end{split}
\end{equation}
(C19)

From Eqs. (C17) and (C19), we see that the wave function in the region of the barrier, $|z| \lesssim l$ is reduced by a factor $l/a_L \ll 1$, as compared to its typical value outside the barrier for $|z| \gtrsim l$.

2. Nonadiabatic channel couplings

We now turn to nonadiabatic channel couplings, which correspond to the terms $C_{na} = -\{p_x, A(x)\}/(2m)$ and $\{p_x, A(z)\} + A^2(z)$ in Eqs. (19) and (48), respectively. In the 2LS, we are interested in the + BO channel. The effective potential in this channel forms a trap as illustrated in Fig. 1(b), whereas the effective potential in the – channel forms an inverted trap. Therefore, the – channel hosts a continuum of scattering states, and the nonadiabatic channel coupling $\{p_x, A(x)\}/(2m)$ induces decay of states in the + channel into the continuum in the – channel. Below, we estimate the corresponding decay rate using Fermi’s golden rule. In the 3LS, we are interested in the zero-energy channel, for which the effective potential is given by the potential barrier in Eq. (50). Decay occurs again to the open – channel.

a. DOS in the open channel

According to Fermi’s golden rule, the rate of decay of a given initial state to an open channel hosting a continuum of final states is determined by the product of the transition matrix element between the initial and final states, and the DOS in the open channel. In this section, we derive the DOS in the open channel, which corresponds to the – BO channel in both the 2LS and the 3LS. We neglect nonadiabatic corrections to the effective potential in the – channel, which is thus determined by Eqs. (15) and (44) for the 2LS and 3LS, respectively. Moreover, for simplicity we consider the symmetric cases $\delta = 1$ and $r^2 = 1$. Then, for both systems, the effective BO potential in the – channel can be written as $-U(x) = -\hbar \sqrt{(\Omega/2)^2 + (\Delta')^2}$, where we use the following identification table:
\begin{equation}
\begin{array}{ccc}
\text{2LS} & \Omega & \Delta' \\
\text{3LS} & \sqrt{2\Omega} & 2\Delta' \\
\end{array}
\end{equation}
(Also, in the 3LS, the $x$ coordinate should be replaced by $z$.) Since we are interested in the resonant decay from energetically higher channels into the – channel, only states with energy $E > -\Omega/2$ are relevant. Such states can be described in the WKB approximation. To write down the corresponding wave functions, we assume that the system is contained in a large box of size $2L$, $|x| \leq L$. (The size $L$ will disappear from the final result for the decay rate.) Then, the normalized wave function can be written as
\begin{equation}
\begin{split}
\psi_{E}^-(x) = \frac{1}{\sqrt{L^{1/4}}} \Phi\left(1, \frac{\Delta'}{\sqrt{E + U(x)}}\right)^{1/4} \\
\times \sin / \cos\left\{\frac{1}{\hbar} \int_{0}^{x} dx' \sqrt{2m(E + U(x))}\right\},
\end{split}
\end{equation}
(C20)
where we neglect terms of order $\Omega/\Delta'L \ll 1$. The choice of $\sin \{\ldots\}$ or $\cos \{\ldots\}$ corresponds to either symmetric or antisymmetric parity of the wave function, respectively. The corresponding eigenenergies $E = E_n$ can be obtained from the WKB quantization condition
\begin{equation}
\begin{split}
\frac{1}{\hbar} \int_{-L}^{L} dx' \sqrt{2mE_n + U(x)} = \pi \left(n + \frac{1}{2}\right).
\end{split}
\end{equation}
(C21)
It follows from this equation that the DOS is given by
\begin{equation}
\frac{dn}{dE} \approx \frac{2\sqrt{2}}{\pi} \sqrt{\frac{mL}{\hbar^3 \Delta'}}
\end{equation}
(C22)
where we neglect terms vanishing for $L \rightarrow \infty$.

b. Decay rate in the 2LS

We now derive the rate of decay from the + BO channel to the – channel in the 2LS. For simplicity, we focus on the symmetric case $\delta = 1$, and we consider the ground state in the harmonic approximation (22) to the effective potential in the + channel as the initial state,
\begin{equation}
\phi_0(x) = \frac{1}{\sqrt{l_0 \sqrt{\pi}}} \exp\left(\frac{x^2}{2l_0^2}\right),
\end{equation}
(C23)
where $l_0 = \sqrt{\hbar/(m\omega_0)}$ and we set $x_0 = 0$. The validity of the harmonic approximation is controlled by the inequality $l_0 \ll s$, where $s$, which is given in Eq. (14), denotes
the width of the resonant region. This inequality will be of crucial importance for the following discussion.

The state \( \phi_0(x) \) in the + channel is coupled to the state described by the wave function \( \psi_E(x) \) in Eq. (C20) in the − channel by the operator \( C_{na} = -\{p_x, A(x)\} / (2m) \), which occurs as a nonadiabatic channel coupling in Eq. (19). Using the aforementioned inequality, this operator can be approximated as

\[
C_{na} = \frac{\hbar^2}{2m} \left[ \frac{s}{s^2 + x^2} \frac{d}{dx} - \frac{sx}{(s^2 + x^2)^2} \right] \begin{pmatrix} 0 & -1 \\ 1 & 0 \end{pmatrix}
\]

(C24)

As a result, the coupling matrix element \( M \) which enters Fermi’s golden rule is

\[
M = \frac{\hbar^2}{2m} \int_{-L}^{L} dx \, \phi_0(x) \begin{pmatrix} \frac{d}{s} dx & -x \\ \frac{s}{s^2} \end{pmatrix} \psi(-E)(x) = M_1 - M_2.
\]

(C25)

Since \( \phi_0(x) \) is a symmetric function of \( x \), the matrix element \( M \) is different from zero only if \( \psi_E(-E)(x) \) is an antisymmetric function. Therefore, one has to choose the \( \sin \{ \ldots \} \) solution in Eq. (C20). Furthermore, one can easily see that the contribution from \( M_2 \) is smaller than that of \( M_1 \) by a factor which is \( \mathcal{O}((l_0/s)^2) \), and thus

\[
M \approx M_1 = \frac{\hbar^2}{2m} \int_{-l}^{l} dx \, \phi_0(x) \frac{d}{dx} \psi_E(-E)(x).
\]

(C26)

The calculation of the above integral can be simplified significantly by using the inequality \( l_0 \ll s \), which yields the following simplified expression of \( \psi_E(-E)(x) \) for \( |x| \sim l_0 \):

\[
\psi_E(-E)(x) \approx \frac{1}{L^{1/4}} \frac{1}{\sqrt{2}} \left[ \frac{\Delta_0^2}{E + \hbar \omega_0/2} \right]^{1/4} \frac{1}{\hbar} x \sqrt{2m[E + \hbar \omega_0/2]} \sin \left( \frac{1}{\hbar} \sqrt{2m} \frac{l_0}{s} \right).
\]

(C27)

Further, we can expand the oscillatory factor, because the neglected terms are much smaller than unity if \( \omega_0/\Omega \ll 1 \). Within this approximation, we obtain for \( E \approx \hbar \omega_0/2 \) the matrix element

\[
M \approx \frac{1}{L^{1/4}} \frac{\hbar^2}{2m} \sqrt{2l_0} \left( \frac{\pi}{2s} \right)^{1/4} \frac{l_0}{s} \exp \left[ -\frac{1}{2} (\tilde{p} l_0)^2 \right].
\]

(C28)

where \( \tilde{p} = \sqrt{4m/\hbar} l_0/\hbar \). This result for the matrix element and the DOS from Eq. (C22) yield the ratio of the decay rate \( \Gamma_{2LS} \) to the oscillator frequency \( \omega_0 \) given in Eq. (23),

\[
\frac{\Gamma_{2LS}}{\omega_0} \approx 2 \sqrt{\pi} \frac{l_0}{s} \exp \left( -8 \left( \frac{s}{l_0} \right)^2 \right).
\]

(C29)

In particular, we find that the decay rate of the + channel for the 2LS is determined by the ratio \( l_0/s \) and is suppressed exponentially for \( l_0 \ll s \).

c. Decay rate in the three-level system

We proceed to calculate the decay rate of the zero-energy BO channel in the 3LS. As discussed in Sec. C1, for \( a_L > l \) the nonadiabatic potential barrier has a strong influence on the wave function in the zero-energy channel. However, to estimate the decay rate, we neglect the effects of the barrier on the open-channel wave function \( \psi_E(-E)(x) \). This is legitimate because the height of the barrier \( \hbar^2/(2m^2) \) is much smaller than the gap \( \Omega/\sqrt{2} \) between the − channel and the zero-energy channel. Therefore, we can use Eq. (C20) with \( E = 0 \) for the final wave function \( \psi_E(0)(x) \) in the open channel,

\[
\psi_E(0)(x) \approx \frac{1}{L^{1/4}} \frac{1}{\sqrt{2}} \left[ \frac{\Delta'}{E-(-\omega_0)} \right]^{1/4} \times \sin \cos \left( \frac{1}{\hbar} \int_0^z d\zeta \sqrt{-2mE_-(\zeta)} \right)
\]

\[
= \frac{1}{L^{1/4}} \frac{1}{\sqrt{2}} \left( \frac{\Omega}{1 + \pi^2} \right)^{1/8} \times \sin \cos \left( \sqrt{\frac{\Omega}{\omega_L a_L}} \int_0^z d\zeta (1 + \pi^2)^{1/4} \right),
\]

(C30)

where \( z = z/l \) and \( E_-(\zeta) \) is defined in Eq. (44). For simplicity we consider the symmetric case with \( r_L^2 = 1 \), for which \( l = \Omega/(\Delta'_\sqrt{2}) \) holds. The nonadiabatic channel coupling \(-\{p_x, A(z)\}/(2m)\) from Eq. (48), with

\[
A(\pi) = \frac{1}{\sqrt{2} \left( 1 + \frac{\pi^2}{2} \right)} \begin{pmatrix} 0 & 1 & 0 \\ 0 & 1 & 0 \end{pmatrix},
\]

(C31)

is nonzero only inside the barrier for \( |z| \lesssim l \), where the behavior of the wave function in the zero-energy channel is governed by the universal solution in Eq. (C7) with \( \lambda = 0 \),

\[
\varphi_0(\tau) = \sqrt{1 + \frac{\pi^2}{2}} \left( A + B \arctan(\tau) \right).
\]

(C32)

The coefficients \( A \) and \( B \) in the above expression depend on details of the behavior of the wave function outside the barrier. We can estimate these coefficients as follows: The typical value of the wave function localized in a spatial area of size \( a_L \) is \( \varphi \sim 1/\sqrt{a_L} \); Further, according to our discussion in Sec. C1, the wave function within the barrier is reduced by a factor of \( l/a_L \). This yields the estimate \( A \sim B \sim l/3 a_L^3/2 \). In the considered case \( l/a_L \gg \sqrt{\omega_L/\Omega} \), for the nonzero coupling matrix element we have

\[
M \approx \frac{\hbar^2}{2m^2} \frac{1}{\sqrt{2} \left( \frac{\Omega}{\omega_L a_L} \right)^{1/4}} \left( \frac{l_0}{s} \right)^{1/4} \cos \left( \sqrt{\frac{\Omega}{\omega_L a_L}} \int_0^z d\zeta (1 + \pi^2)^{1/4} \right),
\]

(C33)
where we keep only the dominant term with the derivative of the open-channel wave function and only differentiate the oscillating term. After performing the standard WKB evaluation of this integral we obtain the estimate

\[ M \approx \frac{\hbar^2}{ma_L^{3/2}} \sqrt{\frac{2}{\sqrt{\Omega}} \frac{1}{\omega L a_L}} \exp \left(-\sqrt{2} \frac{\sqrt{\Omega}}{\omega L a_L} I \right), \]

where

\[ I = \int_0^1 dy (1 - y^2)^{1/4} = \frac{\sqrt{2\pi^{3/2}}}{6\Gamma(3/4)} \approx 0.874. \]

An analogous calculation shows that the channel coupling \( A^2(z) \) yields a matrix element which is smaller than \( M \) in Eq. (C34) by a factor of order \( \mathcal{O}(l/a_L) \sqrt{\omega L/\Omega} \) and thus negligible. With the matrix element Eq. (C34) and the DOS from Eq. (C22), using Fermi’s golden rule we obtain for the ratio of the decay rate \( \Gamma_{3LS} \) to the oscillator frequency \( \omega_L \), the result reported in Eq. (51),

\[ \frac{\Gamma_{3LS}}{\omega_L} \approx 2 \frac{l^2}{a_L^2} \sqrt{\frac{\Omega}{\omega_L}} \exp \left(-2^{5/4} \frac{\sqrt{\Omega}}{\omega_L a_L} I \right) \]

\[ \approx 2 \frac{l^2}{a_L^2} \sqrt{\frac{\Omega}{\omega_L}} \exp \left(-2 \sqrt{\frac{\Omega}{\omega_L a_L}} I \right). \]

We note that the above derivation requires the following hierarchy of scales

\[ 1 \gg \frac{l}{a_L} \gg \sqrt{\frac{\omega_L}{\Omega}}, \]

for which \( \Gamma_{3LS}/\omega_L \) is exponentially suppressed.

**Appendix D: Dipole-dipole matrix elements in the rotating frame**

Here we explicitly calculate the temporal average of the dipole-dipole matrix elements in the rotating frame from Eq. (58). Expanding \( \bar{d}_{2}^{\tilde{N}, \tilde{N}'} \) in the subset \( \{ \tilde{0}, \tilde{1}, \tilde{1}, \tilde{2}, \tilde{0} \} \) of eigenvectors of the rigid rotor in an electric field, see Eq. (4), gives

\[ \bar{d}_{2}^{\tilde{N}, \tilde{N}'} = \sum_{N,N',M,M'} \langle \tilde{N}, 0 | \otimes | \tilde{M}, 0 \rangle \bar{d}_{2}^{\tilde{N}, \tilde{N}'} \langle M', 0 | \otimes \langle N', 0 |. \]

The matrix elements are given by

\[ \bar{d}_{2}^{\tilde{N}, \tilde{N}'} = \lim_{T \to \infty} \frac{1}{T} \int_{-T}^{T} dt \langle \tilde{N}, 0 | \otimes | \tilde{N}, 0 \rangle U_{3LS}^{(1)}(t) | \tilde{M}, 0 \rangle \otimes | \tilde{M}, 0 \rangle \]

\[ \approx \langle \tilde{N}, 0 | \otimes | \tilde{N}, 0 \rangle \langle \tilde{M}, 0 | \otimes | \tilde{M}, 0 \rangle \delta_{\tilde{N}, \tilde{N}'} \delta_{M, M'} \]

\[ = \langle \tilde{N}, 0 | \otimes | \tilde{N}, 0 \rangle \langle \tilde{M}, 0 | \otimes | \tilde{M}, 0 \rangle \delta_{\tilde{N}, \tilde{N}'} \delta_{M, M'} (1 - \delta_{\tilde{N}, \tilde{M}}), \]

where we used that \( U_{3LS}^{(j)}(t) \) is diagonal, see Eq. (10).

Further, the oscillation frequencies are identified by \( \omega_0 = -\omega_1, \omega_1 = 0 \) and \( \omega_2 = \omega_2 \).

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