High-fidelity quantum memory realized via Wigner crystals of polar molecules*

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(Received 13 August 2011; revised manuscript received 5 September 2011)

The collective excitations of spin states of an ensemble of polar molecules are studied as a candidate for high-fidelity quantum memory. To avoid the collisional properties of the molecules, they are arranged in dipolar crystals under one or two dimensional trapping conditions. We calculate the lifetime of the quantum memory by identifying the dominant decoherence mechanisms and estimating their effects on gate operations when a molecular ensemble qubit is transferred to a microwave cavity.

Keywords: polar molecules, quantum memory, molecular dipolar crystals

PACS: 03.67.Lx, 33.80.–b, 85.25.Cp **DOI:** 10.1088/1674-1056/21/1/010308

1. Introduction

Photons are good carriers to transmit quantum information. However, photons can only be used in a probabilistic way[1,2] for quantum information processing and are hard to store. Therefore, many applications in quantum information processing call for reversible and efficient mapping of nonclassical photon states onto electronic excitations in atom[3,4] or solids.[5−8] The development of scalable quantum memory elements is a critical step toward realizing large-scale quantum technology.[9] A quantum memory is challenging to implement, as it must be capable of preserving both the amplitude and the phase of a qubit during the transfer and storage. Here we study such a scenario, in which a superconducting microwave cavity[10] is coupled to a cloud of cold polar molecules serving as a quantum memory with a long coherence time.

As a candidate for quantum memory, polar molecules[11−16] can provide two features: first, the long coherent time for qubits stored in the polar molecules, which is based on identifying long-lived electron and nuclear spin states in the electronic and vibrational ground state manifold; second, the strong coupling of molecular qubits to the microwave cavity, which is based on the transitions between spin excitations with large electric dipole moments of the order of a few Debyes. A qubit stored in the collective spin excitation of a thermal ensemble of N cold polar molecules is coupled to the cavity with an enhanced collective coupling $g_N = g\sqrt{N}$, which however introduces a dephasing of the qubit due to the state-dependent collisions. To completely avoid the collisional properties of the separated molecules, the idea is to arrange them in a crystal similar to the Wigner crystal of trapped ions.[17]

A molecular dipolar crystal is based on electric dipole moments $\boldsymbol{\mu}_{\rm ind}$ of the polar molecules induced by an external direct-current (DC) field, which gives an interaction of $|\boldsymbol{\mu}_{\rm ind}|^2/4\epsilon_0 r^3 \equiv C_3/r^3$ between two molecules at distance r.[13,15] For molecules confined to one dimension (1D) by an external trapping potential and dipole moments aligned perpendicularly, the interaction is repulsive and allows the formation of a high-density crystal, where molecules perform small oscillations around their equilibrium positions, reminiscent of the Wigner crystal of trapped ions.

The stability of such a dipole crystal requires a large ratio between the interaction energy and the kinetic energy, $\Gamma_{\rm d} = U_{\rm int}/E_{\rm kin} \gg 1$, where $\mu_{\rm ind} = |\boldsymbol{\mu}_{\rm ind}|$, $U_{\rm int} = \mu_{\rm ind}^2/4\pi\epsilon_0 R_0^3$, $E_{\rm kin} = \min\{\hbar^2/mR_0^2, k_{\rm B}T\}$, $\hbar$ is the reduced Plank constant, $k_{\rm B}$ is the Boltzmann constant, ϵ_0 is the vacuum permittivity, T is the tempera-

*Project supported by the National Natural Science Foundation of China (Grant Nos. 11004029 and 11174052), the Natural Science Foundation of Jiangsu Province, China (Grant No. BK2010422), the Ph. D. Program Foundation of Ministry of Education of China, the Excellent Young Teachers Program of Southeast University, China, and the Major State Basic Research Development Program of China (Grant No. 2011CB921203).

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ture and R_0 is the mean molecule separation. For the one-dimensional (1D) case, condition $\Gamma_{\rm d} \gg 1$ typically leads to a mean molecule separation of $R_0 \leq 100$ nm and an induced dipole moment of $\mu_{\rm ind} \sim 1$ Debye. For the 2D case, $\Gamma_{\rm d} \gg 15$, we should have a mean molecule separation of $R_0 \leq 50$ nm and and an induced dipole moment of $\mu_{\rm ind} \sim 1$ Debye.

An operational definition of the decoherence time of an ensemble qubit can be given in terms of an idealized experiment. A cavity qubit $\rho_{\rm c}(t=0) = |\psi\rangle_{\rm c}\langle\psi|$ with $|\psi\rangle_{\rm c} = \alpha|0\rangle_{\rm c} + \beta|1\rangle_{\rm c}$ (here $|0\rangle_{\rm c}$ and $|1\rangle_{\rm c}$ represent the cavity states with 0 and 1 photon, respectively) is written at time $t=0$ to the molecular memory with all other molecules initialized in state $|0\rangle$ with a perfect swap operation. The qubit is kept in storage for a time interval τ undergoing dephasing collisions. At $t=\tau$, the qubit is transferred back to the cavity mode, resulting in a reduced density matrix $\rho_{\rm c}(\tau)$ of the cavity with a fidelity $F_\tau = {}_{\rm c}\langle\psi|\rho_{\rm c}(\tau)|\psi\rangle_{\rm c}$. The decoherence time of the ensemble memory is identified as the decay time of the fidelity.

In this paper, we investigate the storage of quantum information encoded in collective excitations (ensemble qubits of spin degrees of freedom in a self-assembled dipolar crystal of polar molecules), which is proved to be protected from the decoherence caused by the dipole–dipole interactions. We study the dynamics of ensemble spin qubits in a self-assembled molecular dipolar crystal, in which the qubits interact with the phonon modes of the crystal. We calculate the lifetime of the quantum memory by identifying the dominant decoherence mechanisms and estimating their effects on the gate operations when a molecular ensemble qubit is transferred to a cavity QED. We study specific setups and conditions, with which the coupling to the phonon modes can be minimized. Detailed results are presented for a one-dimensional dipolar chain. The paper is organized as follows. We start in Section 2 with a definition of the molecular ensemble qubits and the dispersion relation of the system. In Section 3, we study the dynamics of the collective spin excitations in a dipolar crystal due to the state-dependent dipole–dipole interactions and the coupling to the motional degrees of freedom. We derive a general model for the system, calculate the limitation for the lifetime of the ensemble qubits stored in the spin degrees of freedom and show how we can realize a highly protected quantum memory. A summary is given in Section 4.

2. Ensemble qubits

2.1. Definitions

The $|\downarrow\rangle$ and $|\uparrow\rangle$ denote a pair of eigenstates of the Hamiltonian for a molecule, $\hat{H}_{\rm m} = B\hat{N}^2 + \gamma\hat{N}\cdot\hat{S}$, where $\hat{N}$ is the angular momentum of the nuclei, B is the rotational constant, γ is the spin-rotation coupling constant and $\hat{S}$ is the electron spin. The ensemble qubits are shown in the forms of $|0_{\rm e}\rangle = |\downarrow\cdots\downarrow\rangle$, and $|1_{\rm e}\rangle = (1/\sqrt{N})\sum_{i=1}^{N}|\downarrow\cdots\uparrow_i\cdots\downarrow\rangle$. We choose two spin states from calcium monofluoride (CaF), $|\downarrow\rangle = |N=1, J=3/2, M_J=1/2\rangle$ and $|\uparrow\rangle = |N=1, J=3/2, M_J=-1/2\rangle$, then $\mu_{\rm ind} = \langle\downarrow'|\hat{\mu}|\uparrow'\rangle = \langle\uparrow'|\hat{\mu}|\uparrow'\rangle \simeq 0.2\mu_0$ (0.6 Debye) for a field strength of $E_z\mu_0 \simeq 1.9B$, where $|\downarrow'\rangle$ and $|\uparrow'\rangle$ denote the eigenstates of a rigid rotor Hamiltonian

$$\hat{H}_{\rm m} = B\hat{N}^2 + \gamma\hat{N}\cdot\hat{S} - E_z\hat{\mu}_z \tag{1}$$

for a molecule in the present of a bias field $\boldsymbol{E} = E_z\boldsymbol{e}_z$, μ_0 is the vacuum permeability, and $\hat{\mu}_z$ is the dipole operator in the z direction.

2.2. Properties

We consider an ensemble of N polar molecules of mass m (for both 1D and 2D cases). The Hamiltonian is described by

$$\hat{H} = \sum_j^N \frac{\hat{p}_j^2}{2m} + V(\hat{x}_j) + \hat{H}_{\rm m}^{(j)} + \hat{H}_{\rm dd}, \tag{2}$$

where

$$\hat{H}_{\rm dd} = \frac{1}{2}\sum_{j\neq l}\frac{\boldsymbol{\mu}_j\cdot\boldsymbol{\mu}_l - 3(\boldsymbol{\mu}_j\cdot\boldsymbol{e}_x)(\boldsymbol{\mu}_l\cdot\boldsymbol{e}_x)}{\left|\hat{R}_j - \hat{R}_l\right|^3}. \tag{3}$$

We assume that the dominating energy scale is given by the internal states, so we first look at the eigenstates of $\hat{H}_m^{(j)}$. With assumption $E_z\mu_0 \sim \gamma \ll B$, we diagonalize $\hat{H}_m^{(j)}$ in the basis $\{|N, M_N\rangle|S, M_S\rangle\}$ and find that the two eigenstates $|\downarrow'\rangle$ and $|\uparrow'\rangle$ corresponding to the zero field states $|\downarrow\rangle$ and $|\uparrow\rangle$ have the same induced dipole moment for a field strength. We define $\mu_{\rm ind}^{\downarrow} = \langle\downarrow'|\hat{\mu}|\downarrow'\rangle$ and $\mu_{\rm ind}^{\uparrow} = \langle\uparrow'|\hat{\mu}|\uparrow'\rangle$. We take these two states $|\downarrow_i\rangle$ and $|\uparrow_i\rangle$ as our molecular qubit states, where index i labels the i-th molecule. Within the qubit subspace of the two molecules, we decompose the dipole operator into

$$\boldsymbol{\mu}_j\cdot\boldsymbol{\mu}_l - 3(\boldsymbol{\mu}_j\cdot\boldsymbol{e}_x)(\boldsymbol{\mu}_l\cdot\boldsymbol{e}_x) = \mu_{\rm ind}^2(I_{jl} + \hat{K}_{jl}) \tag{4}$$

to single out the contribution that is independent of the qubit state. We choose such $\mu_{\rm ind}^{\downarrow}$ and $\mu_{\rm ind}^{\uparrow}$ that $\langle\downarrow_j,\downarrow_l|\,\hat{K}_{jl}\,|\downarrow_j,\downarrow_l\rangle=0$, where

$$\begin{aligned}\hat{K}_{jl} &= \Big(\frac{\mu_{\rm ind}^{\uparrow}}{\mu_{\rm ind}^{\downarrow}}-1\Big)\big(\,|\downarrow_j',\uparrow_l'\rangle\langle\downarrow_j',\uparrow_l'|+\text{h.c.}\big)\\ &\quad+\left[\Big(\frac{\mu_{\rm ind}^{\uparrow}}{\mu_{\rm ind}^{\downarrow}}\Big)^2-1\right]|\uparrow_j',\uparrow_l'\rangle\langle\uparrow_j',\uparrow_l'|\\ &=\kappa(\sigma_j^-+\sigma_l^+ +\text{h.c.}),\end{aligned}\tag{5}$$

with $\kappa=|\langle 1|\,\boldsymbol{u}\,|0\rangle|^2/\mu_{\rm ind}^2$, and σ being the Pauli operators. Note that if both qubit states have exactly the same dipole moment, $\hat{K}_{jl}=0$.

We now assume that the dipole repulsion and the external trapping potential order the molecule in a crystal with equilibrium positions R_j^0 and small fluctuations around these positions $\hat{r}_j=\hat{R}_j-R_j^0$. Therefore, we expand $\hat{H}_{\rm dd}$ around the equilibrium positions and obtain

$$\begin{aligned}\hat{H}_{\rm dd}^{(0)} &= \frac{1}{2}\frac{\mu_{\rm ind}^2}{4\pi\varepsilon_0}\sum_{j\neq l}\frac{1}{|R_j^0-R_l^0|^3}\\ &\quad+\frac{1}{2}\frac{\mu_{\rm ind}^2}{4\pi\varepsilon_0}\sum_{j\neq l}\frac{1}{|R_j^0-R_l^0|^3}\hat{K}_{jl},\\ \hat{H}_{\rm dd}^{(1)} &= -\frac{3}{2}\frac{\mu_{\rm ind}^2}{4\pi\varepsilon_0}\sum_{j\neq l}\frac{R_l^0-R_j^0}{|R_l^0-R_j^0|^5}\\ &\quad\times(\hat{r}_j-\hat{r}_l)\hat{K}_{jl},\\ \hat{H}_{\rm dd}^{(2)} &= \frac{12}{2}\frac{\mu_{\rm ind}^2}{4\pi\varepsilon_0}\sum_{j\neq l}\frac{(\hat{r}_j-\hat{r}_l)^2}{|R_l^0-R_j^0|^5}\\ &\quad+\frac{12}{2}\frac{\mu_{\rm ind}^2}{4\pi\varepsilon_0}\sum_{j\neq l}\frac{(\hat{r}_j-\hat{r}_l)^2}{|R_l^0-R_j^0|^5}\hat{K}_{jl}.\end{aligned}\tag{6}$$

The first terms of $\hat{H}_{\rm dd}^{(0)}$ and $\hat{H}_{\rm dd}^{(2)}$ together with $\sum_j(\hat{p}_j^2/2m)+V(\hat{r}_j)$ describe the harmonic oscillations of the phonons around the equilibrium positions. The second term of $\hat{H}_{\rm dd}^{(0)}$ describes the interaction between the internal states and determines the energy band of the internal states. The second term of $\hat{H}_{\rm dd}^{(2)}$ can be neglected. The $\hat{H}_{\rm dd}^{(1)}$ describes the interaction between the internal states and the phonon modes. Hence, the Hamiltonian can be rewritten as

$$\begin{aligned}\hat{H} &= \sum_q\omega_q\hat{a}_q^{\dagger}\hat{a}_q+\sum_j^N\hat{H}_{\rm m}^{(j)}+\frac{1}{2}\frac{\mu_{\rm ind}^2}{4\pi\varepsilon_0}\sum_{j\neq l}\frac{\hat{K}_{jl}}{|R_j^0-R_l^0|^3}\\ &\quad-\frac{3}{2}\frac{\mu_{\rm ind}^2}{4\pi\varepsilon_0}\sum_{j\neq l}\frac{R_l^0-R_j^0}{|R_l^0-R_j^0|^5}(\hat{r}_j-\hat{r}_l)\hat{K}_{jl}.\end{aligned}\tag{7}$$

The last term describes a state-dependent force on the molecules and entangles the internal and the external degrees of freedom.

2.3. Energy band

2.3.1. The 1D case: a homogeneous chain

In this section, we consider the energy band of a molecular ensemble with N molecules prepared in a crystalline phase and the qubits encoded in the collective rotational excitations. We start with the simplest possible model of a homogeneous 1D crystal. We obtain the dispersion relation

$$\omega_q^2=\frac{6\mu_{\rm ind}^2}{\pi\varepsilon_0 mR_0^5}\sum_{j=1}^{N}\frac{1}{j^5}\sin^2\frac{jq}{2}.\tag{8}$$

In the limit of large N and small q, the spectrum of the acoustic phonon modes in the dipolar crystal is of the from

$$\omega_q\approx\omega_0 q=\sqrt{6\zeta(3)\frac{\mu_{\rm ind}^2}{4\pi\varepsilon_0R_0^3}\frac{1}{mR_0^2}}\,q,\tag{9}$$

where ζ is the Riemann zeta function.

The frequency of the ensemble qubit in the 1D homogeneous chain is obtained as

$$\begin{aligned}\Omega_k^{\rm 1D} &= \frac{1}{2\hbar}\frac{\mu_{\rm ind}^2}{4\pi\varepsilon_0}\langle\bar{k}|\sum_{j\neq l}\frac{1}{|R_j^0-R_l^0|^3}\hat{K}_{jl}\,|\bar{k}\rangle\\ &\simeq\frac{1}{\hbar}\frac{9\mu_{\rm ind}^2}{4\pi\varepsilon_0R_0^3}\zeta(3)\Delta\mu,\end{aligned}\tag{10}$$

where $|\bar{k}\rangle=(1/\sqrt{N})\sum_j\mathrm{e}^{\mathrm{i}kj}\,|\downarrow\cdots\uparrow_j\cdots\downarrow\rangle$, $k=2\pi n/N$, $n=0,\ldots,N-1$, and $\Delta\mu=\mu_{\rm ind}^{\uparrow}/\mu_{\rm ind}^{\downarrow}-1$.

2.3.2. The 2D case: a regular triangular lattice

Now we consider the energy band of the molecular ensemble with dipolar molecules in a 2D lattice. A 2D regular triangular lattice is used as an example. With the boundaries of the first Brillouim zone in the momentum space of the triangular lattice, we obtain the two modes of the phonon spectrum along a path through three points $\Gamma(0,0)$, $M(0,\frac{\pi}{a}\frac{2}{\sqrt{3}})$, and $K(\frac{\pi}{a}\frac{2}{3},\frac{\pi}{a}\frac{2}{\sqrt{3}})$.

In the limit of large N and small q, the phonon spectra are

$$\begin{aligned}\omega_{q_x}&\approx\omega_0 q_x=1.38\sqrt{\frac{6\mu_{\rm ind}^2}{\pi\varepsilon_0a^3}\frac{1}{ma^2}}\,q_x,\\ \omega_{q_y}&\approx\omega_0 q_y=1.53\sqrt{\frac{6\mu_{\rm ind}^2}{\pi\varepsilon_0a^3}\frac{1}{ma^2}}\,q_y.\end{aligned}\tag{11}$$

The frequency of the ensemble qubit in the 2D regular triangular lattice is obtained as

$$\Omega_k^{2\mathrm{D}} = \frac{1}{2\hbar}\frac{\mu_{\mathrm{ind}}^2}{4\pi\varepsilon_0}\langle\bar{k}|\sum_{j\neq l}\frac{1}{\left|R_j^0-R_l^0\right|^3}\hat{K}_{jl}\left|\bar{k}\right\rangle$$
$$= \frac{9\Delta\mu}{2\hbar}\frac{\mu_{\mathrm{ind}}^2}{4\pi\varepsilon_0 a^3}\sum_{j_x,l_x,j_y,l_y}\frac{1}{[(j_x-l_x)^2+(j_x-l_x)(j_y-l_y)+(j_y-l_y)^2]^{3/2}}. \quad (12)$$

3. Decoherence

In this section, we consider the dominant source of decoherence of the ensemble qubit. As ensemble qubit state $|1_{\mathrm{e}}\rangle$ is almost an eigenstate of $\hat{H}_{\mathrm{dd}}^{(0)}$, thus the error is dominated by the thermal fluctuation provided by $\hat{H}_{\mathrm{dd}}^{(1)}$. For a molecular ensemble in a harmonic trap with irregular mean equilibrium positions, the error is introduced during both the holding time (dominated by $\hat{H}_{\mathrm{dd}}^{(1)}$) and the gate time (dominated by $\hat{H}_{\mathrm{dd}}^{(0)}$).

3.1. The regular lattice

First, we consider the decoherence of the ensemble qubit with molecules trapped in the regular lattice. The error during the holding time is dominated due to $\hat{H}_{\mathrm{dd}}$.

For the 1D regular lattice, we consider the interaction between the external and the internal states introduced by the dipole–dipole interaction $\hat{H}_{\mathrm{dd}}$ in the interaction picture:

$$\hat{H}_{\mathrm{int}}^{1\mathrm{D}} = -\frac{3}{2}\frac{\mu_{\mathrm{ind}}^2}{4\pi\varepsilon_0}\sum_{j\neq l}\frac{R_l^0-R_j^0}{\left|R_l^0-R_j^0\right|^5}(\hat{x}_j-\hat{x}_l)\hat{K}_{jl}$$
$$= -\frac{3}{2}\frac{\mu_{\mathrm{ind}}^2}{4\pi\varepsilon_0 R_0^3}\sqrt{\frac{\hbar}{2mR_0^2}}\sum_{k,k'}\sum_q\frac{f^{1\mathrm{D}}(k,k',q)\delta(k-k'+q)}{\sqrt{\omega_q}}\left|\bar{k}'\right\rangle\langle\bar{k}|\left[(\hat{a}_q^\dagger \mathrm{e}^{\mathrm{i}\omega_q t}+\hat{a}_q\mathrm{e}^{-\mathrm{i}\omega_q t})\right], \quad (13)$$

where

$$f^{1\mathrm{D}}(k,k',q) = 9\Delta\mu\left(\mathrm{Li}_4\left(\mathrm{e}^{\mathrm{i}(k-k')}\right)-\mathrm{Li}_4\left(\mathrm{e}^{-\mathrm{i}(k-k')}\right)-\mathrm{Li}_4(\mathrm{e}^{\mathrm{i}q})+\mathrm{Li}_4(\mathrm{e}^{-\mathrm{i}q})\right), \quad (14)$$

and Li is the polylogarithm function.

For molecules trapped in the 2D regular lattice (here a triangle lattice is used as an example), the Hamiltonian of the dipole–dipole interactions between the molecules is obtained as

$$\hat{H}_{\mathrm{int}}^{2\mathrm{D}} = -\frac{3}{2}\frac{\mu_{\mathrm{ind}}^2}{4\pi\varepsilon_0}\sum_{j\neq l}\frac{R_l^0-R_j^0}{\left|R_l^0-R_j^0\right|^5}(\hat{r}_j-\hat{r}_l)\hat{K}_{jl}$$
$$= -\frac{3}{2}\frac{\mu_{\mathrm{ind}}^2}{4\pi\varepsilon_0 a^3}\sqrt{\frac{\hbar}{2ma^2}}\sum_{k,k'}\Bigg\{\sum_{q_x}\frac{f^{2\mathrm{D}}(k,k',q_x)\delta(k-k'+q_x)}{\sqrt{\omega_{q_x}}}\left|\bar{k}'\right\rangle\langle\bar{k}|\left[(\hat{a}_{q_x}^\dagger \mathrm{e}^{\mathrm{i}\omega_{q_x}t}+\hat{a}_{q_x}\mathrm{e}^{-\mathrm{i}\omega_{q_x}t})\right]\boldsymbol{e}_x$$
$$+\sum_{q_y}\frac{f^{2\mathrm{D}}(k,k',q_y)\delta(k-k'+q_y)}{\sqrt{\omega_{q_y}}}\left|\bar{k}'\right\rangle\langle\bar{k}|\left[(\hat{a}_{q_y}^\dagger \mathrm{e}^{\mathrm{i}\omega_{q_y}t}+\hat{a}_{q_y}\mathrm{e}^{-\mathrm{i}\omega_{q_y}t})\right]\boldsymbol{e}_y\Bigg\}, \quad (15)$$

where

$$f^{2\mathrm{D}}(k,k',q) = 9\Delta\mu\sum\frac{2\cos k\left[(j_x-l_x)+(j_y-l_y)\right]}{\left[(j_x-l_x)^2+(j_x-l_x)(j_y-l_y)+(j_y-l_y)^2\right]^2}. \quad (16)$$

For the zero temperature case, at $t=0$, the initial state is $|i\rangle = |1_{\mathrm{e}}\rangle|0\rangle_p^{\otimes N}$. The probability of molecules in state $|f\rangle = \left|\bar{k}\right\rangle|0\cdots 1_k\cdots 0\rangle_p$ at time t is obtained by using Fermi's golden rule (we only consider the molecular ensemble trapped in the 1D lattice as an example)

$$P_f^{1\mathrm{D}}(0) = \frac{1}{\omega_{\mathrm{m}}}\int_0^{\omega_{\mathrm{m}}}\mathrm{d}\omega\left|\frac{i}{\hbar}\int_0^t\mathrm{d}t'\langle f|H_{\mathrm{int}}(t')|i\rangle\,\mathrm{e}^{\mathrm{i}(E_f-E_i)t'/\hbar}\right|^2$$
$$= \frac{1}{\omega_{\mathrm{m}}}\int_0^{\omega_{\mathrm{m}}}\mathrm{d}\omega\left[\frac{1}{\hbar^2}\left(\frac{3}{2}\frac{\mu_{\mathrm{ind}}^2}{4\pi\varepsilon_0 R_0^3}\sqrt{\frac{\hbar}{2mR_0^2}}\right)^2\frac{1}{\omega}\left|f^{1\mathrm{D}}(0,k,k)\right|^2\frac{2\sin^2(\omega t/2)}{\omega^2}\right]$$

$$\simeq \frac{\sqrt{6\zeta(3)}}{\pi}\sqrt{\Gamma_d}\Delta\mu^2 F(\omega_{\rm m}t), \tag{17}$$

where

$$F(\omega_{\rm m}t) = \frac{1}{2}\left[\gamma + \ln(\omega_{\rm m}t) - {\rm Ci}(\omega_{\rm m}t)\right]. \tag{18}$$

If $\omega_{\rm m}t < 1$, $F(\omega_{\rm m}t) \simeq 0.12(\omega_{\rm m}t)^2$. Whereas if t is large enough, then $F(\omega_{\rm m}t) \simeq \frac{1}{2}\left[\gamma + \ln(\omega_{\rm m}t)\right]$. For final temperature T case,

$$\begin{aligned} P_f^{\rm 1D}(T) &= \frac{1}{\omega_{\rm m}}\int_0^{\omega_{\rm m}} {\rm d}\omega \left[\frac{1}{\hbar^2}\left(\frac{3}{2}\frac{\mu_{\rm ind}^2}{4\pi\varepsilon_0 R_0^3}\sqrt{\frac{\hbar}{2mR_0^2}}\right)^2 \frac{1}{\omega}\left|f^{\rm 1D}(0,k,k)\right|^2 \frac{2\sin^2(\omega t/2)}{\omega^2}\left[\bar{n}(\omega)+1\right]\right] \\ &= \frac{\sqrt{6\zeta(3)}}{\pi}\sqrt{\Gamma_d}\Delta\mu^2 G(\omega_{\rm m}t), \end{aligned} \tag{19}$$

where

$$G(\omega_{\rm m}t) = F(\omega_{\rm m}t) - \frac{1}{2}\frac{k_{\rm B}T}{\hbar\omega_{\rm m}}\sin^2\left(\frac{1}{2}\omega_{\rm m}t\right) + \frac{1}{4}\frac{k_{\rm B}T}{\hbar}t{\rm Si}(\omega_{\rm m}t), \tag{20}$$

and $\bar{n}(\omega) = ({\rm e}^{\hbar\omega/k_{\rm B}T} - 1)^{-1}$.

Hence we can calculate the fidelity of the initial state of the ensemble qubit during the holding time, $\mathcal{F}_{\rm h} = 1 - P_{\rm f}$. For an ensemble qubit stored in the spin states of the molecules, the coherent time is not limited by the long range dipole–dipole interactions, as $\Delta\mu \ll 1$, we should include the effect from the stray of the static electric field $\partial(\mu_{\rm ind}^{\uparrow} - \mu_{\rm ind}^{\downarrow})/\partial E_z$. For example, $|\downarrow\rangle = |N=1, J=3/2, M_J=1/2\rangle$ and $|\uparrow\rangle = |N=1, J=3/2, M_J=-1/2\rangle$, $\partial(\mu_{\rm ind}^{\uparrow} - \mu_{\rm ind}^{\downarrow})/\partial E_z$ is about $10^{-6}\mu_0$ as shown in Fig. 1.

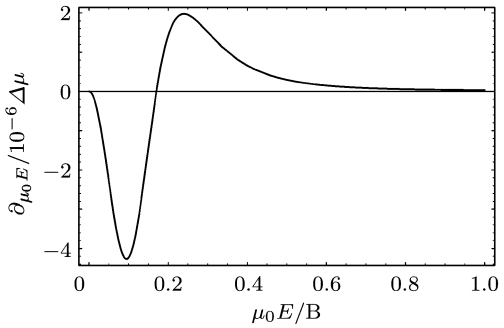


Fig. 1. The $\partial(\mu_\uparrow - \mu_\downarrow)/\partial(\mu_0 E)$ of spin states versus applied external electric field strength.

During a gate operation in a three-level system, $|\downarrow\rangle$ and $|\uparrow\rangle$ are coupled by a Raman process involving a single cavity photon and an external microwave field, and $|r\rangle$ is an excited state, we obtain an additional effective interaction of the form

$$\hat{H}_{\rm eff} = U_{\rm eff}\frac{R_0^3}{2}\sum_{i\neq j}\frac{|\downarrow_i\rangle\langle\uparrow_j| + |\uparrow_i\rangle\langle\downarrow_j|}{|\hat{R}_i - \hat{R}_j|^3}, \tag{21}$$

where $U_{\rm eff} = g_{\rm m}^2|\langle\downarrow|\hat{\mu}_z|r\rangle|^2/4\pi\varepsilon_0 R_0^3\Delta^2$, and Δ is the detuning from excited state $|r\rangle$. The $g_{\rm m}$ is an effective cavity–molecule coupling given in the form of

$$g_{\rm m} = \frac{\sqrt{N}|\langle\downarrow|\hat{\mu}_z|r\rangle|E_{\rm c}\Omega(t)}{2\hbar\Delta}, \tag{22}$$

where $E_{\rm c} \simeq \sqrt{\hbar\omega_{\rm c}/2\pi\varepsilon_0 d^2 L}$ is the electric field per photon for a cavity of length $\simeq$ 1 cm and typical electrode distance $d \simeq 10$ μm, $\Omega(t) = |\langle\uparrow|\hat{\mu}_{x-y}|r\rangle|E_{\rm dr}(t)/\hbar \approx 2g_N$ is the controllable Rabi frequency of the external microwave field with the frequency of the driving field being $E_{\rm dr}$, and $\omega_{\rm c}$ is the frequency of the cavity.. After operation time $\tau \approx g_{\rm m}^{-1}$, the gate error is $\varepsilon \approx \langle\Delta\hat{H}_{\rm eff}^2\rangle/4\Delta^2$. And the fidelity of the gate is obtained for the polar molecules in the 1D regular chain as

$$\begin{aligned} \mathcal{F}_{\rm g}^{\rm 1D} &\equiv 1 - \Delta H_{\rm eff}^2/4\Delta^2 \\ &\approx 1 - \Big(\frac{1}{\hbar}\frac{|\langle\downarrow|\hat{\mu}_z|r\rangle|^2}{4\pi\varepsilon_0 R_0^3}\Big)^2\frac{1}{4\Gamma_d\Delta^2}. \end{aligned} \tag{23}$$

Here we give an example. With electric dipole matrix elements $|\langle\uparrow|\hat{\mu}_{x-y}|r\rangle| \simeq 0.05\mu_0$ and $|\langle\downarrow|\hat{\mu}_z|r\rangle| \simeq 0.5\mu_0$, the strength of the driven field $\mu_0 E_{\rm dr} \simeq$ 10–100 MHz, $N = 10^4$–10^6, $R_0 = 100$ nm, the induced dipole moment $\mu_{\rm ind} \simeq 0.6$ Debye (for CaF), and the mass of a single molecule $m \simeq 1.7\times10^{-25}$ kg, we obtain a coupling strength of 1–10 MHz, and the fidelity of the gate is $\mathcal{F}_{\rm g}^{\rm 1D} \geq 0.998$.

3.2. An irregular lattice

To study polar molecules trapped in a 1D or 2D irregular lattice, we consider a chain of N molecules in a trap. The molecules are assumed to be strongly bound in the y and the z directions while weakly bound in a harmonic potential in the x direction.

First, we show the deocherence during the swap gate operation and the fidelity of the gate operation.

The fidelity of the gate is obtained as

$$\mathcal{F}_{\rm g}^{\rm irr} \approx 1 - \Big(\frac{1}{\hbar}\frac{|\langle\downarrow|\hat{\mu}_z|r\rangle|^2}{4\pi\varepsilon_0 R_0^3}\Big)^2 \frac{f(N)}{4\Delta^2}, \qquad (24)$$

where $f(N)$ is a function of N, which can be solved either analytically with some approximations (see below for details) or numerically as shown in Fig. 2.

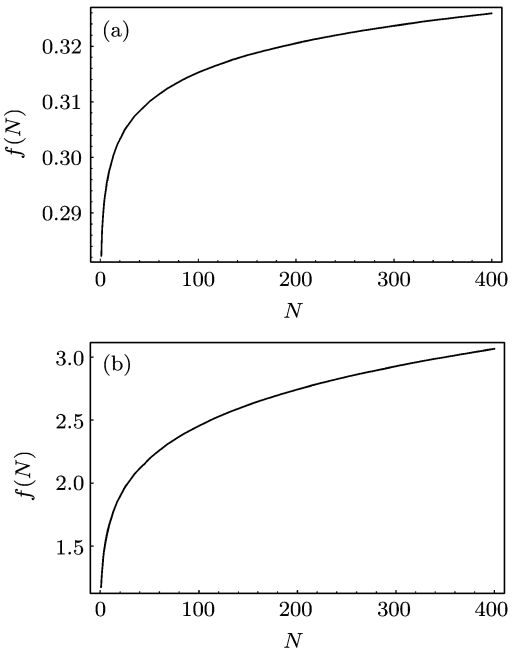


Fig. 2. Numerical results of $f(N)$ for polar molecules trapped in (a) a 1D irregular lattice and (b) a 2D irregular lattice.

Here we give an example. For fixed minimum spacing of the molecules, R_0, the fidelity of the gate is $\mathcal{F}_{\rm g}^{\rm irr}$. We consider the worst cases: $f(N) < 0.36$ for 1D, $f(N) < 6$ for 2D. We obtain $\mathcal{F}_{\rm g}^{\rm irr} > 0.999$ for 1D with the parameters of $|\langle\downarrow|\hat{\mu}_z|r\rangle| \simeq 1.5$ Debye, $R_0 \simeq 100$ nm, and $\Delta \simeq 10$ MHz. For the 2D case, we obtain $\mathcal{F}_{\rm g}^{\rm irr} > 0.999$ with the parameters of $|\langle\downarrow|\hat{\mu}_z|r\rangle| \simeq 0.15$ Debye, $R_0 \simeq 50$ nm, and $\Delta \simeq 10$ MHz (here $g_{\rm m} \simeq 100$ kHz–1 MHz).

Our goal in the following is to study the properties of excitons (dipole moments) and phonons in a polar molecular ensemble in the presence of an additional harmonic confinement potential and compare them with those of the homogeneous crystal shown in Section 3. Although in the inhomogeneous case, we cannot derive exact analytic expressions for those quantities, good approximate solutions can be found in the limit of large N. The derivations of the following results are based on a similar calculation in Ref. [18] for the phonon spectrum of a harmonically confined ion crystal.

3.2.1. Density of molecules in trap

In the first step, we need to determine the equilibrium positions of the N dipoles confined by the harmonic trapping potential. The position of the m-th molecule numbered from left to right will be denoted $x_{\rm m}$. The motion of each molecule will be influenced by the harmonic potential and the dipole–dipole interaction. Hence, the potential energy of the molecule chain is given by

$$V = \frac{1}{2}m\nu^2 \sum_{m=1}^{N} x_{\rm m}^2 + \frac{1}{2}\frac{\mu_{\rm ind}^2}{4\pi\varepsilon_0}\sum_{m\neq n}\frac{1}{|x_{\rm m}-x_n|^3}, \qquad (25)$$

where m is the mass of a molecule, and ν is the trap frequency, which characterizes the strength of the trapping potential in the axial direction.

Assume that the ions are sufficiently cold and that the position of the m-th molecule can be approximated by

$$x_m \approx x_m^{(0)} + q_m, \qquad (26)$$

where $x_m^{(0)}$ is the equilibrium position of the molecule, and q_m is a small displacement. The equilibrium positions will be determined by

$$\left[\frac{\partial V}{\partial x_m}\right]_{x_m = x_m^{(0)}} = 0. \qquad (27)$$

If we define the length scale l by the formula

$$l^5 = \frac{\mu_{\rm ind}^2}{8\pi\varepsilon_0}\frac{1}{m\nu^2} \qquad (28)$$

and dimensionless equilibrium position by $u_m = x_m^{(0)}/l$, then equation (27) may be rewritten as the following set of N coupled algebraic equations of u_m:

$$u_m - 3\sum_{n=1}^{m-1}\frac{1}{(u_n-u_m)^4} + 3\sum_{n=m+1}^{N}\frac{1}{(u_n-u_m)^4} = 0. \qquad (29)$$

For large values of N, it is necessary to solve u_m numerically as shown in Fig. 3.

By inspection, we find that the minimum value of the spacing between two adjacent molecules occurs at the centre of the chain. Compiling the numerical data for the minimum value of the separation for different numbers of trapped molecules, we find that it obeys the following relation:

$$u_{\rm min}(N) \approx \frac{1.914}{N^{0.396}}. \qquad (30)$$

Thus, the minimum inter-molecule spacing for different numbers of molecules is given by $lu_{\rm min} = \left(\frac{\mu_{\rm ind}^2}{8\pi\varepsilon_0}\frac{1}{m\nu^2}\right)^{1/5}\frac{1.914}{N^{0.396}}$.

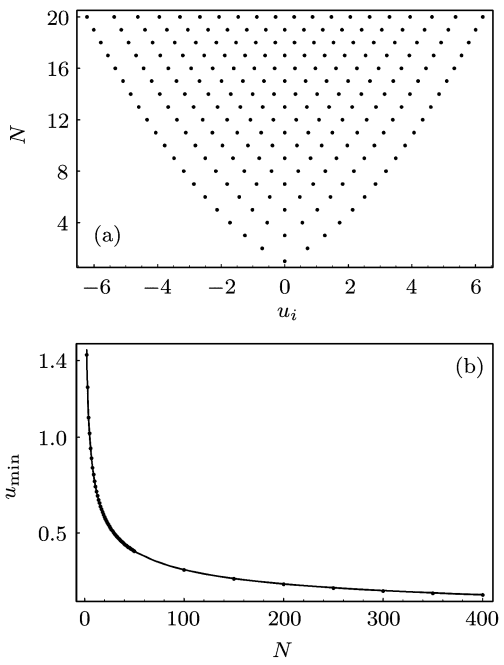


Fig. 3. (a) Equilibrium positions as a function of N. (b) Relationship between the number of trapped molecules N and the minimum separation. The curve is given by Eq. (30) and the dots come from the numerical solutions of Eq. (29).

Using fixed minimum distance $R_0 = 100$ nm, the induced dipole moment $\mu_{\rm ind} \simeq 0.6$ Debye (for CaF), the mass of a single molecule $m \simeq 1.7 \times 10^{-25}$ kg and $N = 10^4$, we obtain that the trap frequency is about 50 Hz and the length of the crystal is about 1.2 mm.

A systematic derivation of an analytic solution to the eigenstates of the dipole–dipole interaction Hamiltonian shown in Eq. (21) is a challenge, since it requires systematically taking into account the position-dependent coupling constant and the long-range interaction. Nevertheless, in the limit of a large number of molecules, $N \gg 1$, we can make some simplifying assumptions. In this limit, the interparticle spacing $a_L(x_i) = x_{i+1}^{(0)} - x_i^{(0)}$ is a smooth function of the equilibrium positions, and it is inversely proportional to the number of molecules in a unit length,

$$n_L(x_i) = \frac{1}{a_L(x_i)}. \tag{31}$$

One goal will be to attain some analytic results on density $n_L(x_i)$. We will construct an approximate expression for energy U of the system, which depends on $n_L(x_i)$. The functional minimization of U with respect to $n_L(x_i)$ will determine the variation of the density.

Suppose that the molecule chain is infinitely long and homogeneous in x. The energy of this homogeneous system can be determined by $U^{\rm hom}/N = \frac{1}{2}\frac{\mu_{\rm ind}^2}{4\pi\varepsilon_0} n^3 \zeta(3)$, where n is the density of the molecules. Provided that n is slowly varying, we can employ Eq. (31) to find U for the nonuniform system in the local density approximation

$$U(n) = \frac{1}{2} m\nu^2 \int n(x)\, x^2 \mathrm{d}x + \frac{1}{2}\frac{\mu_{\rm ind}^2}{4\pi\varepsilon_0}\zeta(3) \int n^4(x)\, \mathrm{d}x. \tag{32}$$

As

$$\begin{aligned} &\delta U(n) \\ &= \int \mathrm{d}x \delta n(x) \left[\frac{1}{2} m\nu^2 x^2 + \frac{\mu_{\rm ind}^2}{4\pi\varepsilon_0} 2\zeta(3)\, n^3(x) - c\right] \\ &\equiv 0, \end{aligned} \tag{33}$$

we obtain

$$n(x) = \frac{5\Gamma(5/6)\, N}{2\sqrt{\pi}\Gamma(1/3)\, L}\left(1 - \frac{x^2}{L^2}\right)^{1/3} \tag{34}$$

for $|x| \le L$ with the numerical constant $\Gamma(5/6)/\Gamma(1/3)\sqrt{\pi} = 0.24$, and $n(x) = 0$ for the other cases, where $2L$ is the length of the crystal at equilibrium. So we have $\int_{-L}^{L} n(x)\, \mathrm{d}x = N$. Length L is evaluated by minimizing the energy of the crystal,

$$L = \left[\frac{5\Gamma(5/6)\, \zeta^{1/3}(3)\, N}{\sqrt{\pi}\Gamma(1/3)}\right]^{3/5} l. \tag{35}$$

For simplification, we can also expand $n(x)$ to the 4-th order with the trial variational function $n(x) = A - Bx^2 - Cx^4$ for $|x| \le L$ and $n(x) = 0$ for the other cases. The resulting 1D density is

$$n(x) = \frac{15N}{26L}\left(1 - \frac{x^2}{3L^2} - \frac{x^4}{9L^4}\right). \tag{36}$$

Length L is obtained as $L = \left[(15/13)\zeta^{1/3}(3)\, N\right]^{3/5} l$. From Eq. (36), we obtain the minimum value of the separation, $u_{\min} = (26/15)^{2/5} (8\zeta(3))^{1/5} N^{-0.4}$, which agrees with the numerical simulation result Eq. (30) well (Fig. 4).

Equations (35) and (36) express the dependence of the centre density and the length of the crystal on molecule number N. For comparison with the case of a homogeneous crystal discussed above, it is more convenient to fix the density at the trap centre $n(0)$.

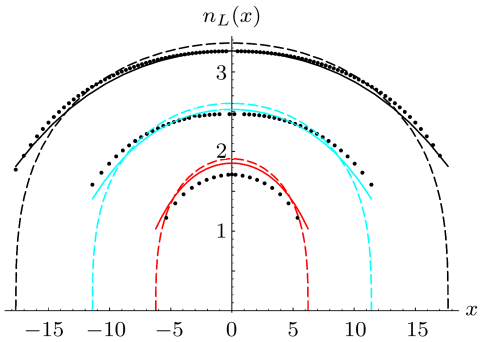


Fig. 4. (colour online) Density n as a function of position x. The dots, the dashed lines and the solid lines give the results obtained by the numerical simulation of Eq. (34) and Eq. (36), respectivly. The red, the blue and the black denote the results obtained with $N = 20,\ 50,\ 100$, respectively. Both x and $n_L(x)$ are scaled to l.

3.2.2. Eigenstates of the dipole–dipole interaction Hamiltonian in the long-wavelength limit

We define the rescaled positions $\xi_i^{(0)} = x_i^{(0)}/L$ and the rescaled interparticle distances $a(\xi_i) = a_L(x_i)/L$. With these definitions, the n-th eigenstate of the Hamiltonian (or $H_{\rm rd}$) shown in Eq. (21) takes the form

$$|\varphi\rangle_n = \sum_i c_i^{(n)} |\downarrow \cdots \uparrow_i \cdots \downarrow\rangle , \tag{37}$$

with eigenenergy E_n. The coefficients of the state obey the relation

$$U_{\rm eff} \frac{R_0^3}{2} \sum_{i \neq j} \frac{c_i^{(n)}}{\left| x_i^{(0)} - x_j^{(0)} \right|^3} = E_n c_j^{(n)}. \tag{38}$$

The coefficients are shown in Fig. 5. If the number of the molecules is large ($N \gg 1$), for small n, we can approximate the chain by a continuous distribution of molecules. In this limit, ξ is a continuous variable varying in the interval of $(-1, 1)$, and the coefficient is a continuous function, which will be denoted as $c(\xi)$. Then Eq. (37) takes the form

$$I(\xi, c(\xi)) = \widetilde{E}_n c(\xi), \tag{39}$$

where

$$I(\xi, c(\xi)) = \int_{-1}^{\xi - a(\xi)} \frac{n(\xi')}{(\xi - \xi')^3} c(\xi')\, \mathrm{d}\xi' + \int_{\xi + a(\xi)}^{1} \frac{n(\xi')}{(\xi' - \xi)^3} c(\xi')\, \mathrm{d}\xi', \tag{40}$$

and $n(\xi) = \left(1 - \xi^2\right)^{1/3}$ is the density of molecules normalized to $5\Gamma(5/6)/[2\sqrt{\pi}\Gamma(1/3)]$. Equations (39) and (40) are valid away from the edges of the chain, where the continuum approximation is reasonable.

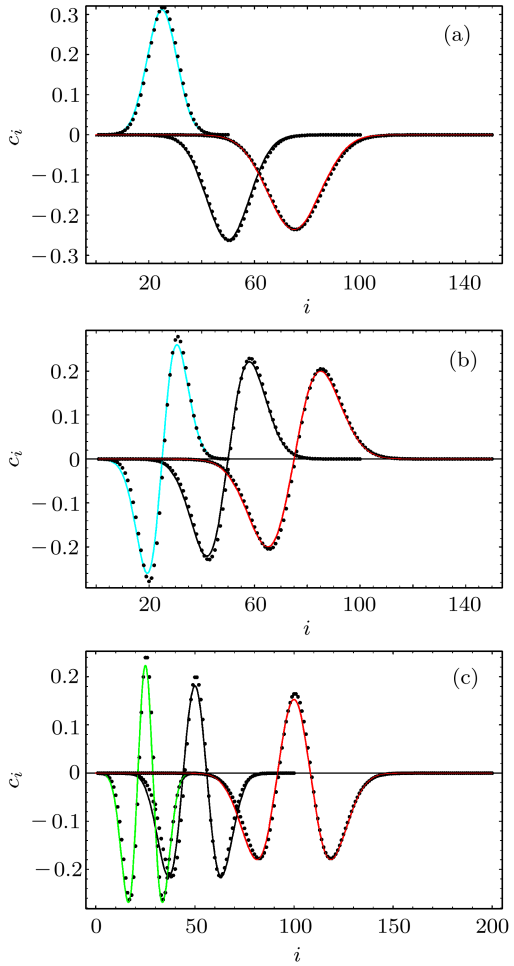


Fig. 5. (colour online) Coefficients of (a) the first (most symmetric), (b) the second, and (c) the third eigenstates of the dipole–dipole interaction Hamiltonian, where c_i is a function of index i. The dots denote the results determined by numerical simulation, the lines denote the analytical results in the long-wavelength limit. The blue, the black and the red denote the results obtained with $N = 50,\ 100,\ 150$, respectively.

By the partial integration, Eq. (40) can be written as the sum of two terms

$$I = I_0 + \Delta I, \tag{41}$$

where I_0 contains the contributions of the ions around point ξ, and ΔI is determined by the values of the density and function $c(\xi)$ and their derivatives at the end-points of the chain. In the long-wavelength limit, at point ξ sufficiently far away from the chain end-points, we find $I \simeq I_0$, and

$$I_0 = \frac{1}{2a(\xi)^2}\left[f(\xi - a(\xi)) + f(\xi + a(\xi))\right] - \frac{1}{2a(\xi)}\left[f'(\xi - a(\xi)) - f'(\xi + a(\xi))\right] - \frac{1}{2}\ln a(\xi)\left[f''(\xi - a(\xi)) + f''(\xi + a(\xi))\right]$$

$$+ \mathrm{O}\left(a\left(\xi\right)\right) \simeq \frac{1}{a^2\left(\xi\right)} f f''\left(\xi\right), \tag{42}$$

with $f(\xi) = n(\xi)c(\xi)$. The coefficients of the eigenstate reduce to

$$c''\left(\xi\right) + \frac{2n''\left(\xi\right)}{n\left(\xi\right)} c'\left(\xi\right) + \left[\frac{n''\left(\xi\right)}{n\left(\xi\right)} + \left(\frac{5\Gamma\left(5/6\right)}{2\sqrt{\pi}\Gamma\left(1/3\right)}\right)^2 \frac{N^2}{\ln N} n^2\left(\xi\right) - \frac{\widetilde{E}}{n\left(\xi\right)}\right] c\left(\xi\right) = 0. \tag{43}$$

There are two independent even and odd solutions of Eq. (43),

$$c_{\mathrm{e}}\left(\xi\right) = \frac{\mathrm{e}^{-\sqrt{b}\xi^2}}{\left(1-\xi^2\right)^{1/3}} {}_1\mathrm{F}_1\left(-\frac{a-\sqrt{b}}{4\sqrt{b}}, \frac{1}{2}, \sqrt{b}\xi^2\right),$$
$$c_{\mathrm{o}}\left(\xi\right) = \frac{\mathrm{e}^{-\sqrt{b}\xi^2}}{\left(1-\xi^2\right)^{1/3}} {}_1\mathrm{F}_1\left(-\frac{a-\sqrt{b}}{4\sqrt{b}} + \frac{1}{2}, \frac{3}{2}, \sqrt{b}\xi^2\right), \tag{44}$$

where $a = (\frac{5\Gamma(5/6)}{2\sqrt{\pi}\Gamma(1/3)})^2 \frac{N^2}{\ln N} - \widetilde{E}$, $b = \frac{2}{3}(\frac{5\Gamma(5/6)}{2\sqrt{\pi}\Gamma(1/3)})^2 \frac{N^2}{\ln N} + \widetilde{E}/3$, and ${}_1\mathrm{F}_1$ is the Kummer confluent hypergeometric function. The $\widetilde{E}_{\mathrm{e}}$ and $\widetilde{E}_{\mathrm{o}}$ are the eigenenergies, which can be obtained by boundary conditions ${}_1\mathrm{F}_1\left(-(a-\sqrt{b})/(4\sqrt{b}), 1/2, \sqrt{b}\right) = 0$ and ${}_1\mathrm{F}_1\left(-(a-\sqrt{b})/(4\sqrt{b}) + 1/2, 3/2, \sqrt{b}\right) = 0$, respectively. The first two orders of eigenenergies are obtained in the form

$$\widetilde{E}_0 = \frac{1}{6}\left[-11 + 6\left(\frac{5\Gamma(5/6)}{2\sqrt{\pi}\Gamma(1/3)}\right)^2 \frac{N^2}{\ln N} - \sqrt{-23 + 36\left(\frac{5\Gamma(5/6)}{2\sqrt{\pi}\Gamma(1/3)}\right)^2 \frac{N^2}{\ln N}}\right],$$
$$\widetilde{E}_1 = \frac{1}{6}\left[-9 + 2\left(\frac{5\Gamma(5/6)}{2\sqrt{\pi}\Gamma(1/3)}\right)^2 \frac{N^2}{\ln N} - 3\sqrt{-7 + 4\left(\frac{5\Gamma(5/6)}{2\sqrt{\pi}\Gamma(1/3)}\right)^2 \frac{N^2}{\ln N}}\right]. \tag{45}$$

Note that the total width of the exciton spectrum is exactly the same as that in the regular lattice case. This is due to the fact that the long-wavelength excitons are located at the centre of the trap, where the density is almost a constant.

Although our derivation is based on several rather crude approximations, we find by comparing with the numerical results that our analytic solutions for E and c can provide an accurate description of the long-wavelength behaviour of the exciton spectrum and the shape of the eigenfunctions. Due to the similarity of the underlying equations, this approach should also be applicable to improve the linear phonon spectrum obtained in Ref. [18] for a harmonically confined ion crystal.

3.2.3. State transfer fidelities for spin-ensemble qubits

For a simplified discussion of the state transfer fidelity between the microwave cavity and the spin-ensemble qubits, we identify the gate fidelity $\mathcal{F}_{\mathrm{g}}^{\mathrm{irr}}$ with the probability to convert a single cavity photon into a spin excitation. The fidelity is degraded through two processes. First, the spin state decays due to the finite admixture of the rotational ensemble state. For a detuning Δ and the fast gate time considered in this paper, this decay is quadratic and the corresponding rate is $f(N)$. Second, the photon state decays linearly with cavity decay rate Γ_{c}. Assuming that each of the two processes acts for approximately half of the gate time, we obtain a total gate fidelity

$$\mathcal{F}_{\mathrm{g}}^{\mathrm{irr}} \simeq 1 - \left(\frac{1}{\hbar}\frac{\pi|\left\langle\downarrow\right|\hat{\mu}_z\left|r\right\rangle|^2}{4\pi\varepsilon_0 R_0^3}\right)^2 \frac{\mathcal{I}_{\mathrm{exc}}}{4\Delta^2} - \frac{\pi\Gamma_{\mathrm{c}}\Delta}{4g_N^2}, \tag{46}$$

where $\mathcal{I}_{\mathrm{exc}}$ is proportional to the exciton dispersion, which arises in an irregular lattice from the mismatch between the cavity mode function and the exciton eigenfunctions. In terms of the normalized exciton spectrum $\bar{E}_n = (E_n - \hbar\omega_{\mathrm{eg}})/\Delta H_{\mathrm{eff}}$ and the overlap $z_n = \sum_i c_i^{(n)} u(x_i)$, it is given by $\mathcal{I}_{\mathrm{exc}} = \sum_n \bar{E}_n^2 z_n^2 - \left(\sum_n \bar{E}_n z_n^2\right)^2$. For cavity mode function $u(x) = \cos(\pi x/\lambda_{\mathrm{c}})$ and $L \leq \lambda_{\mathrm{c}}/2$, we obtain $\mathcal{I}_{\mathrm{exc}} \simeq 0.3$, where the lower value corresponds to $L = \lambda_{\mathrm{c}}/2$.

4. Conclusion

In this paper, we have investigated the storage of quantum information using collective excitations (molecular ensemble qubits of long-lived spin degrees of freedom in a self-assembled dipolar crystal of polar molecules), which provides a high-fidelity quantum memory that can be coupled to the microwave cavity of a solid-state quantum processor. We demonstrate the potential of the molecule dipolar crystals in the context of hybrid quantum computing, since high gate fidelities and long storage time can be combined with the gate time.

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