Franck–Condon physics in a single trapped ion

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Abstract. We propose a way to explore the Franck–Condon (FC) physics using a single ion confined in a spin-dependent potential, formed by the combination of a Paul trap and a magnetic field gradient. A correlation between electronic and vibrational degrees of freedom, called electron–vibron coupling, is induced by a non-zero gradient. In the case of a sufficiently strong electron–vibron coupling, FC blockade of low-lying vibronic transitions takes place. We examine the feasibility of observing FC physics in a single trapped ion and demonstrate many potential applications of ionic FC physics in quantum state engineering and quantum information processing.

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1. Introduction

The Franck–Condon (FC) principle is a well-known fundamental law for explaining the intensity of vibronic transitions in molecules [1, 2], in which the transition intensity is proportional to the FC factor defined by the square of the overlap integral between the vibrational wavefunctions of the two involved states. The FC physics actually exists in various systems of interaction between mechanical and electronic degrees of freedom. In particular, a very small or zero FC factor will cause transition suppression, called the FC blockade, and a non-zero FC factor between different vibrational modes may cause vibrational sidebands [3, 4].

Besides the conventional experiments on molecules, the electronic transport through quantum dots could be exponentially suppressed in the region of strong electron–vibron coupling [5]. The conspicuous transport property in the strong coupling regime plays an important role in both single-molecule devices [6] and nano-electromechanical systems [7].

To the best of our knowledge, the FC physics has still not been clearly demonstrated in a true single-particle system. A recent experiment with an ensemble of atoms confined within a spin-dependent optical lattice [8] for sideband cooling and coherent operations via FC physics can be regarded as an effective single-particle implementation if the double occupation or the inter-atom interaction is negligible. However, as the system is cooled to only populating the ground and first-excited vibrational states for each lattice cell, the s-wave scattering between atoms in a realistic experiment is unavoidable if double occupation appears. For a realistic system of shallow lattices, the atoms can easily tunnel between neighboring sites, leading to the possibility of multiple particles in a particular site. For a realistic system of deep lattices, if the atomic number per lattice site is larger than one, the on-site interaction between atoms cannot be completely ignored. Even for a system without inter-particle interaction, due to the intrinsic nature of a spin-dependent optical lattice, the unavoidable coupling between next-nearest-neighboring sites may destroy the FC physics and induce complex quantum transport along the lattice axis. For example, if the potential regarding the atomic lower level is proportional to \( \sin^2(kx) \) (with \( k \) being the wave number and \( x \) the lattice axis) and the potential relevant to the atomic upper level is proportional to \( \sin^2(kx + \phi) \) (with \( \phi \) being a phase shift), the atom in the lattice site at \( x = 0 \) may jump to the left site at \( x = -\frac{\phi}{k} \) or to the right site at \( x = \frac{1}{k} (\pi - \phi) \) with spin inversion. In particular, the probabilities for jumping to these two sites are almost the
same if $\phi \to \frac{\pi}{2}$. Moreover, due to the wavelength and intensity limits of the optical lattices, the maximum shift and the total number of vibronic states are both limited.

In this paper, we present a proposal for observing the FC physics using a true single-atom system, i.e. a single trapped ion, and discuss the potential applications of FC physics in quantum state manipulation. The key point is that the electron–vibron coupling in a single trapped ion is induced and controlled by a magnetic field gradient (MFG). Compared with the electron–vibron coupling generated by radiation of non-resonant laser beams on the ion, which is too weak to observe the FC physics, the MFG-induced electron–vibron coupling is controllable and can be strong enough to observe. We may apply this coupling to suppress or even block some undesired transitions, called FC blockade, or to enhance some desirable transitions. Attributed to its clear environment and high controllability, a single trapped ion opens up a new horizon for study of the FC physics at the single-atom level. Beyond the fundamental interest in various fields ranging from quantum spectroscopy to quantum transport, the ionic FC physics has promising applications in quantum state engineering.

2. The model and the Franck–Condon blockade

We consider a single ultracold ion confined in a Paul trap [9] and the ion only populates two possible electronic levels $|\downarrow\rangle$ and $|\uparrow\rangle$ of different magnetic dipole moments. In the usual ion trap in the absence of MFG, radiation of laser beams on the ion with blue- and red-detuning could yield couplings between the vibrational and electronic degrees of freedom. But this coupling is generally weak for the ultracold ion within the Lamb–Dicke limit (LDP). For ensuring FC blockade between low-lying vibrational states, an MFG with a sufficiently large gradient is required to enhance the electron–vibron coupling. Without loss of generality, for a one-dimensional trap and the gradient along the axis, the electron–vibron coupling could be expressed as

$$H_0 = g\mu_b b \cdot \frac{\sigma_z}{2} \cdot \delta z = G \cdot \frac{\sigma_z}{2} (a^\dagger + a), \quad (1)$$

with the electron-spin $g$-factor, the Bohr magneton $\mu_B$ and the phonon creation (annihilation) operators $a^\dagger$ ($a$). $\delta z$ is the oscillation amplitude of the ion along the $z$-axis. Here, $b$ denotes the MFG sensed by the ion, and $G = g\mu_b b \sqrt{\hbar/2m\omega_z}$ with the ion mass $m$ and the trap frequency $\omega_z$.

The spin-dependent potentials can be written as

$$V_\sigma = H_0 + \frac{1}{2}m\omega_z^2 z^2 = \frac{1}{2}m\omega_z^2 (z + z_0 \sigma_z)^2, \quad (2)$$

and is sketched in figure 1. With the electron–vibron coupling given in equation (1), it is easy to obtain the shift $z_0 = \frac{\mu_b b}{m\omega_z^2}$. For a large gradient $b$, $z_0$ would be sufficiently large and the overlap between the low-lying vibrational wavefunctions becomes very small. As a result, the transition between the two vibrational ground states, $|0, \downarrow\rangle \leftrightarrow |0, \uparrow\rangle$, would be strongly suppressed. This is the FC blockade.

Specifically, due to the non-zero gradient, the ionic electron–vibron coupling is governed by a new effective LDP $\eta' = \sqrt{\eta^2 + \epsilon^2}$, where $\eta$ is the original LDP in the case of no gradient, and $\epsilon = \frac{\omega_0}{\hbar} \sqrt{\hbar/2m\omega_z}$ with $\frac{\omega_0}{\hbar} = \frac{2\pi}{\hbar} b$ is the additional LDP caused by the gradient [10, 11]. This additional LDP has been observed in a recent experiment [12].
Figure 1. Schematic diagram of the spin-dependent trap for a single ion. The red solid and blue dashed curves are the potentials for the ion in $|\uparrow\rangle$ and $|\downarrow\rangle$, respectively. Here, $|n\rangle$ where $n=0, 1, 2$ denotes the vibrational levels. For a sufficiently large distance between the two equilibrium positions, there is no significant overlap between the two low-lying vibrational states so that the FC blockade appears.

Baker–Campbell–Hausdorff formula $e^{A+B} = e^A e^B e^{-1/2[A,B]}$ (under the condition $[A, [A, B]] = [B, [A, B]] = 0$), the transition matrix elements [4], [13–15] between the vibrational states $|0, \sigma\rangle$ and $|n, \sigma'\rangle$ ($\sigma, \sigma' = \downarrow, \uparrow$) could be written as

$$M_{0 \rightarrow n} = M_{|0, \sigma\rangle \leftrightarrow |n, \sigma'\rangle} = \langle n| e^{i\eta'(a+a^\dagger)}|0\rangle = e^{\frac{\eta'^2}{2}} \langle n| \sum_{j,k} (i\eta')^{j+k} j!k! a^j (a^\dagger)^k |0\rangle = e^{\frac{-\eta'^2}{2}} (i\eta')^n \sqrt{n!}. \quad (3)$$

Thus the FC factor is $|M_{0 \rightarrow n}|^2 = e^{-\eta'^2} \frac{\pi^{2n}}{n!}$, which is exponentially sensitive to the ionic electron–vibron coupling, as plotted in figure 2. By adjusting the trap frequency $\omega_z$ and/or the gradient $b$, the effective LDP $\eta'$ will suppress the transition between vibrational ground states of different electronic levels, while allowing transitions between the vibrational ground and excited states of different electronic levels. As shown in figure 2, the maximum FC factors gradually decrease with increasing phonon number $n$. However, the FC factors between different vibrational states remain non-zero, which may cause the vibrational sidebands that are useful for cooling the ion.

3. Observation of Franck–Condon blockade via controlled-NOT gates

The FC physics in a single trapped ion could be observed using a blockade-induced controlled-NOT (CNOT) gate. To this end, we encode the control (target) qubits in the vibrational (internal) states of the ion. For the carrier transitions between the states $|n, \downarrow\rangle$ and $|n, \uparrow\rangle$, we have the Rabi frequency

$$\Omega_{n,n} = \frac{1}{\hbar} \langle n, \uparrow| H_1 |\downarrow, n\rangle = \lambda e^{\frac{-\eta'^2}{2}} |L_n(\eta'^2)|. \quad (4)$$
\[ H_I = \hbar \lambda [\sigma_+ e^{i\eta'(a^\dagger a)} + \sigma_- e^{-i\eta'(a^\dagger a^\dagger)}], \]

with the magnetic dipole coupling strength \( \lambda \), \( \sigma_+ = |\uparrow\rangle\langle\downarrow| \), \( \sigma_- = |\downarrow\rangle\langle\uparrow| \) and the Laguerre polynomial \( L_n(x) \).

To encode two of the vibrational states as a control qubit, one may choose a proper gradient to block the low-lying vibrational transition and enable the high-lying vibrational transition. For simplicity, we choose the low-lying vibrational state \( |0\rangle \) to be blocked and calculate the FC factors \( |M_{n\rightarrow n}|^2 \) for transitions \( |n, \sigma\rangle \leftrightarrow |n, \sigma\rangle \) with \( n = 0, 1, 2, 3 \), respectively. See figure 3(a). It indicates that the quantum transitions \( |0, \downarrow\rangle \leftrightarrow |0, \uparrow\rangle \) and \( |1, \downarrow\rangle \leftrightarrow |1, \uparrow\rangle \) are almost completely blocked if \( \eta' > 3 \), while the FC factor for \( |2, \downarrow\rangle \leftrightarrow |2, \uparrow\rangle \) is still not small up to \( \eta' \sim 3.5 \). So in the region of \( 3 < \eta' \leq 3.5 \), we may encode the vibrational states \( |0\rangle \) and \( |2\rangle \) as the control qubit. By driving the carrier transition for a duration \( \tau \) satisfying \( \Omega_{2,2} \tau = \pi/2 \), the states \( |\downarrow\rangle \) and \( |\uparrow\rangle \) are interchanged only if the vibrational state is \( |2\rangle \). Then we could obtain that \( \Omega_{0,0}/\Omega_{2,2} = 1/(1 - 2\eta'^2 + \eta'^4/2) < 0.0425 \) if \( \eta' > 3 \). This means that, comparing to \( \Omega_{2,2} \tau \), \( \Omega_{0,0} \tau \) can be ignored. Therefore, we may construct a CNOT gate in the subspace spanned by \( |0, \downarrow\rangle \), \( |0, \uparrow\rangle \), \( |2, \downarrow\rangle \) and \( |2, \uparrow\rangle \).

\[
U = \begin{pmatrix}
\cos \Omega_{0,0} \tau & i \sin \Omega_{0,0} \tau & 0 & 0 \\
i \sin \Omega_{0,0} \tau & \cos \Omega_{0,0} \tau & 0 & 0 \\
0 & 0 & \cos \Omega_{2,2} \tau & i \sin \Omega_{2,2} \tau \\
0 & 0 & i \sin \Omega_{2,2} \tau & \cos \Omega_{2,2} \tau
\end{pmatrix}.
\]

Eliminating the undesired phase factors on \( |2\rangle \), we obtain a CNOT gate on the electron states conditional on the vibrational states of the ion: \( |0, \downarrow (\uparrow)\rangle \rightarrow |0, \downarrow (\uparrow)\rangle \) and \( |2, \downarrow (\uparrow)\rangle \rightarrow |2, \downarrow (\uparrow)\rangle \). The fidelity of the CNOT gate is estimated with respect to the effective LDP \( \eta' \) in figure 3(b), in which the growth of \( \eta' \) improves the fidelity.
The FC physics is also observable in the mediate region of $2.6 \leq \eta' < 3$, where the FC factor for $|0, \downarrow \rangle \leftrightarrow |0, \uparrow \rangle$ is almost zero, but the FC factors for $|1, \downarrow \rangle \leftrightarrow |1, \uparrow \rangle$ and $|2, \downarrow \rangle \leftrightarrow |2, \uparrow \rangle$ are still significant. Because $|M_{2\rightarrow 2}|^2 > |M_{1\rightarrow 1}|^2$, we may still encode the control qubit in $|0\rangle$ and $|2\rangle$. The CNOT gate could still be expressed by equation (6) and its fidelity versus $\eta'$ is shown in figure 3(b). Compared to the case of larger gradients, an unfavorable effect may appear as a result of the unwanted population on the vibrational state $|1\rangle$. A simple estimation of the detrimental influence from the undesired population on $|1\rangle$ is obtained in figure 3(c) by assuming the initial vibrational state as $[\alpha (|0\rangle + |2\rangle) + \beta |1\rangle]/\sqrt{2}$ with the error factor $\beta$.

When the gradient is tuned into the region $0 < \eta' \leq 1$, the FC blockade does not happen for the ground vibrational transition but for other high-lying vibrational transitions at some special points, such as the blockade of $|1, \downarrow \rangle \leftrightarrow |1, \uparrow \rangle$ at $\eta' = 1$ and the blockade of $|2, \downarrow \rangle \leftrightarrow |2, \uparrow \rangle$ at $\eta' = 0.765$. This reminds us of the ‘magic’ LDP mentioned in [16]. Therefore, tuning the effective LDP to the ‘magic’ point $\eta' = 1$, we may employ the vibrational states $|0\rangle$ and $|1\rangle$ to encode the control qubit. The CNOT gate is then implemented in the subspace spanned by $|0, \downarrow \rangle$, $|0, \uparrow \rangle$, $|1, \downarrow \rangle$ and $|1, \uparrow \rangle$, where the internal states flip only when the vibrational state is $|0\rangle$. Alternatively, tuning the effective LDP to the other ‘magic’ point $\eta' = 0.765$, we may encode the control qubit in the vibrational states $|0\rangle$ and $|2\rangle$. Different from the schemes for large and mediate gradients, our CNOT gate in the region of small gradients can only be accomplished at some ‘magic’ points. As a result, the quality of the performance is very sensitive to the effective LDP. We have estimated this sensitivity and the relations of the MFG to the LDP in figure 4.

In our CNOT proposal, since two motional states of the ion are encoded as the control qubit, it requires ground-state cooling, which could be done before the MFG is applied or in
Figure 4. (a) The required gradient $b$ versus the LDP $\eta'$ for different trap frequencies. (b) Fidelity of CNOT gating versus the fluctuation of the gradient $\Delta b$ in the case of $\omega_z = 2\pi \times 100 \text{kHz}$: red dashed curve for $b = 624 \text{Tm}^{-1}$ (i.e. $\eta' = 3$); blue solid curve for $b = 208 \text{Tm}^{-1}$ (i.e. $\eta' = 1$) and the green dashed dotted curve for $b = 159.2 \text{Tm}^{-1}$ (i.e. $\eta' = 0.765$).

4. Applications in quantum state engineering

4.1. Preparation of Fock states

The observable FC physics could be used to prepare motional Fock states in a probabilistic way. We consider the initial state

$$|\Psi(0)\rangle = |\downarrow\rangle \langle \downarrow| (P_0 |0\rangle \langle 0| + P_1 |1\rangle \langle 1| + P_2 |2\rangle \langle 2|),$$

with the population probability $P_k$ and the average phonon number $P_1 + 2P_2 < 1$. To prepare the ground motional state $|0\rangle$, one has to eliminate the populations in states $|1\rangle$ and $|2\rangle$. By tuning the gradient to block the transition $|1, \downarrow\rangle \leftrightarrow |1, \uparrow\rangle$, the population in $|1\rangle$ could be screened away via a $\pi/2$ carrier transition pulse following a measurement on $|\uparrow\rangle$. Similarly, by tuning the gradient to block the transition $|2, \downarrow\rangle \leftrightarrow |2, \uparrow\rangle$, the population in $|2\rangle$ could be screened away via a $\pi/2$ carrier transition pulse following a measurement on $|\downarrow\rangle$. In the above operations, the generation of the expected Fock state is probabilistic. So we have to employ the repeat-until-success method [17]. Once the desired internal state is successfully detected, the desired motional state is prepared with unity fidelity.

4.2. Modification for single-qubit gate operations

Due to the existence of FC blockade in the regime of large MFG, some new difficulties for single-qubit operations in a string of trapped ions appear. The proposals [10, 11] of Wunderlich’s group focused on the regime of small MFG, which corresponds to a small LDP. Under such a small LDP, the first-order expansion works very well and the ground-state cooling is indeed not necessary. In the regime of higher MFG supporting the FC blockade, the large MFG favors a working Ising coupling within a shorter time, which makes the two-qubit conditional operations
faster. However, since the large MFG strongly suppresses some vibrational transitions, it becomes more difficult to perform the Hadamard gates via carrier transitions.

We show a specific simulation for a spin flip in the presence and absence of MFG in figure 5. In our simulation, the single trapped ion within the initial internal state $|\downarrow\rangle$ and the thermal motional state (e.g. $\langle n \rangle = 5$ or 0.1) under an MFG ($\eta' = 1$) evolves according to the dynamical population

$$P_{\downarrow}(t) = \langle \downarrow | Tr_n [e^{-iH_It/\hbar} \rho(0)|\downarrow\rangle \langle \downarrow | e^{iH_It/\hbar}] |\downarrow\rangle,$$

where \(\rho(0) = \sum_{n=0} M_n |n \rangle \langle n|\), \(M_n = N[\langle n \rangle / (1 + \langle n \rangle)]^n\) and \(N\) is the normalization constant.

Figure 5 shows the necessity to cool the ion to the vibrational ground state in the presence of a large MFG. One possible solution is to increase the Rabi frequency by enhancing the radiation. Since the suppression is sensitive to \(\eta'\), the appropriate Rabi frequency could be determined by interrogative pulse operations with respect to different values of \(\eta'\). This result is also applicable to the refocusing pulses [18] for removing the undesired couplings due to the Ising coupling [19]. As the refocusing pulses are based on carrier transitions, like the Hadamard gate above, stronger pulses are necessary for refocusing under a large MFG. Alternatively, we may switch off the MFG when performing single-qubit gates. To this end, the employment of lasers, instead of microwaves as in [10, 11], is necessary for individually accomplishing the single-qubit operations, where the FC physics does not work in the carrier transition and cooling to the vibrational ground state is unnecessary.

5. Experimental feasibility and challenge

For a real experimental implementation of the CNOT gate, we may employ the hyperfine levels \(|S_{1/2}, F = 0, m_F = 0\rangle\) and \(|S_{1/2}, F = 1, m_F = 1\rangle\) of \(^{171}\text{Yb}^+\) as \(|\downarrow\rangle\) and \(|\uparrow\rangle\), respectively.
Here, the transition frequency for $|\downarrow\rangle \leftrightarrow |\uparrow\rangle$ is $\omega_0 \approx 2\pi \times 12.6$ GHz [10, 20]. With the $z$-axis trap frequency $\omega_z = 2\pi \times 100$ kHz and magnetic dipole coupling strength $\lambda = 2\pi \times 50$ kHz, the CNOT gate will be accomplished within 19.2, 8.2 and 6.7 $\mu$s for $\eta' = 3$, 1 and 0.765, respectively. To perform the CNOT gates highly coherently, we require the coherence time of the employed hyperfine levels to be longer than at least 192 $\mu$s. Fortunately, the latest experiment has shown that this coherence time could be 5 ms.\textsuperscript{5}

To achieve our scheme, moreover, we may employ the weak region of $\eta'$, e.g. $\eta' = 1$. To this end, due to sensitivity to the ‘magic’ points, a highly stable gradient is required for achieving the high-fidelity operations. We have estimated the sensitivity to the fluctuation of the gradient; see figure 4(b) where the fidelities are larger than 0.994 for all schemes if the gradient fluctuation $|\Delta b| \leq 10$ T m$^{-1}$. In comparison to the paper [16] where the ‘magic’ LDP is controlled by the wave vector, phase and intensity of the radiating lasers, in our scheme the CNOT operation is mainly governed by the MFG, whose strength and stability are key to the implementation.

With the currently available technologies, a big challenge for our scheme is how to realize a large MFG. Current-carrying coils in an anti-Helmholtz-type arrangement [10, 21] and permanent magnets [12] have been applied to attain a gradient up to tens of T m$^{-1}$. To achieve a working Ising-type interaction between two ions separated by a few micrometers, the MFG is required to be of the order of 100 T m$^{-1}$ [22, 23]. Although this is still challenging with current techniques, there are some efforts toward this aim using new materials and improved designs [24].

6. Conclusions

In summary, we have explored the FC physics in a single trapped ion under an external MFG and discussed the experimental feasibility. Although our discussion above focused on the FC blockade, the strong electron–vibron coupling would probably be useful for quantum simulation of e.g. the Dirac equation [25] or quantum walk [26]. We argue that our study would be useful in further understanding FC physics and its applications.

Moreover, the FC physics with strong electron–vibron coupling might also be useful in sideband cooling of the trapped ion, in which vibrational sidebands play an important role and the carrier transitions are excluded. In usual schemes, the cooling does not happen if the cooling laser is orthogonal to the trapping direction [27]. However, for an ion in a spin-dependent potential as shown in figure 1, the electron–vibron coupling is caused by the MFG, instead of the cooling laser itself. As a result, the cooling could work even for a red-detuning beam perpendicular to the trapping direction. However, due to the FC blockade, cooling down to the ground motional state is sometimes impossible, but deterministically to certain motional states, e.g., to $n = 1$ for $b = 208$ T m$^{-1}$ with $\nu = 100$ kHz. Intuitively, the cooling efficiency should be enhanced using the conventional laser cooling techniques plus an MFG in parallel. But cooling the trapped ion to the ground vibrational state using conventional laser techniques requires the trap frequency to be of the order of MHz [28], which requires an MFG larger than $b = 208$ T m$^{-1}$ for efficient cooling. In this sense, it would be better to accomplish the cooling of the trapped ion in our scheme before the MFG is applied. Finally, we noticed that there is a recent work on enhanced cooling via MFG [29], in which the FC physics has not been specifically mentioned.

\textsuperscript{5} Private communication with the Ion Trap Group at Siegen University (2011).

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