The role of phases and their interplay in molecular vibrational quantum computing with multiple qubits

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Abstract. Within the scope of molecular quantum computing with vibrational qubits, we analyse the impact of phases that are present during the quantum computation processes. While the phase relation in superposition states and its temporal evolution are crucial to any implementation of quantum computing, we elucidate the special challenge that emerges for phase control of qubits encoded in molecular vibrational eigenstates. Phase correctly prepared superposition states in general exist only for a finite time and with the inherent entanglement in molecular vibrational qubit systems their development displays a complex pattern. We show that the free relative phase evolution in such qubit systems can be utilized for the implementation of quantum phase gates. Moreover, a practical experimental realization of phase correct quantum gates acting on molecular vibrational qubits could be accomplished by a decomposition into laser-induced population transfer and free evolution phase gates. This concept adds to the flexibility in the implementation of quantum gate sequences and algorithms. A modification, where only a reduced number of selected relative phases needs to be adjusted, will make this scheme more robust and versatile. Finally, we also disclose and discuss another key feature for the implementation of phase correct quantum gates, i.e. the dependence of the quantum gate fidelity on the absolute or carrier-envelope phase of the driving femtosecond laserfield.
1. Introduction

The notion of quantum information in general and the search for the ideal system to implement quantum cryptography or a quantum computer have led to exciting research with invaluable results on the behaviour and controllability of quantum systems during the last two decades [1]–[10]. Probably the most intriguing part of this search is devoted to the challenging task of manipulating a quantum system to the extent of population and phase control, necessary for the realization of the quantum gates, i.e. the corresponding unitary transformations.

In the field of molecular quantum computing with vibrational qubits, we have demonstrated by theoretical investigations in various model systems that it is possible to implement a universal set of quantum logic gates by shaped femtosecond laserfields with high fidelities [11]–[18]. Moreover, we have presented a theoretical implementation of a quantum algorithm [14]. For a detailed discussion of this proposal, we refer the reader to [12, 14]. The concept of molecular vibrational quantum computing is lately also being addressed and investigated by other groups [19]–[23].

The task of population control and its dependences on molecular properties has been analysed and discussed extensively in earlier papers [13], [15]–[17]. Methods for optimizing laserfields that induce a phase correct quantum gate, i.e. with respect to a defined standard basis and any corresponding superposition basis, have already been presented and elucidated in [14, 23]. Yet, knowledge about the phase evolution in multi qubit systems subsequent to laser-driven switching processes, i.e. the ‘lifetimes’ of the phase correctly prepared qubit states, is equally crucial for a successful implementation of quantum gate sequences and algorithms. Moreover, the relation between the laserfield phase and the phase development in the qubit-basis has not been investigated so far. In this paper, we want to analyse the role and the consequences of the interplay between these different types of phases for molecular vibrational quantum computing. We present numerical simulations in multi-qubit systems, which point out basic essentials for future experimental realization.

Following a short introduction to the concept of molecular quantum computing and the application of optimal control theory, we first will elucidate the impact of the free phase evolution
on the controllability of multi-dimensional qubit systems. Furthermore, we demonstrate the utilization of free phase evolution in a flexible and robust scheme for implementing phase correct quantum gates as well as quantum algorithms. The last part of this paper is devoted to the role of the absolute or carrier-envelope phase (CEP) of the driving laserfields regarding the fidelity of the desired quantum gates.

2. Molecular vibrational quantum computing

For molecular vibrational quantum computing, the states $|0\rangle$ and $|1\rangle$ of one qubit are encoded in different excitations of one optically addressable vibrational normal mode [11]. In a polyatomic system, there is a maximum number of $3N - 6$ (for nonlinear molecules) vibrational normal modes available for the representation of one qubit respectively. The multi-qubit standard basis states $|q_1\rangle_k \otimes |q_2\rangle_k \otimes |q_3\rangle_k \cdots \otimes |q_n\rangle_k \equiv |(q_1, q_2, q_3, \cdots, q_n)\rangle_k$, where $q_i$ is either 0 or 1, are encoded in vibrational eigenstates of a molecule which correspond to combinations of different excitations of the normal modes.

Vibrational multi-qubit systems differ from other implementation schemes, like atoms or ions in a trap, linear optics or cavity QED, in a significant way: the eigenenergies already include the coupling via the electronic potential between the normal modes that encode the qubits. Consequently, these multi-qubit states cannot be represented as a product basis, i.e. entanglement between them is always present and the qubits are never evolving independently (see also [14]). In case of non-degeneracy each normal mode has a different excitation energy and each $n$-qubit basis state $|q_1, q_2, q_3, \cdots, q_n\rangle_k$ has a different eigenenergy. In addition, for a given qubit $n$, the prevailing coupling gives rise to $2^n - 1$ slightly different excitation frequencies which depend on the values of all other qubits. For example, for the last qubit in an $n$-qubit basis, there are the $2^n - 1$ excitation energies for the transitions $|(q_1, q_2, q_3, \cdots, 0)_m\rangle \leftrightarrow |(q_1, q_2, q_3, \cdots, 1)_m\rangle$ with $q_1, q_2, q_3, \cdots \in \{0, 1\}$ and $m = 1 \cdots 2^{n-1}$.

This shift in excitation energies of multi-qubit states also provides for the straightforward implementation of conditional two qubit gates, without any external coupling force.

The essential quantum gates for universal quantum computing, NOT, controlled-NOT (CNOT), Hadamard and $\Pi_1$ gate can be implemented on the vibrational qubit states by specially shaped, ultrashort laserfields. These laserfields must induce all desired transformations for each state of the multi-qubit standard basis. To address one qubit individually, the corresponding transitions starting from each of the $2^n$ multi-qubit standard basis states have to be selectively driven by the same specially shaped laserfield (for Hadamard gates there are even $2^{n+1}$ transitions).

2.1. Phase and population control applying optimal control theory

We obtain the laserfields that act as global quantum gates on the vibrational qubits by iteration equations which are derived from the maximization of a special multi-target optimal control
functional, adapted to drive multiple transitions by one laserfield $\epsilon(t)$ \cite{12}

$$J (\Psi_{ik}(t), \Psi_{ik}(t), \epsilon(t)) = \sum_{k=1}^{N} \left[ |\tau_k|^2 - C_k (\Psi_{ik}(t), \Psi_{ik}(t), \epsilon(t)) \right] - \int_0^T \alpha(t)|\epsilon(t)|^2 \, dt,$$

(2)

where $\Psi_{ik}(t)$ are the propagated initial qubit states, $\Psi_{ik}(t)$ (Lagrange multipliers) are the propagated target states, $\tau_k$ are the targets, the $C_k$ are the constraints that have to be fulfilled for the individual transitions (i.e. compliance with the time-dependent Schrödinger equation $\{-\frac{i\bar{\hbar}}{\hbar} [\hat{H}_0 - \tilde{\mu}\epsilon(t)] + \frac{\partial}{\partial t}\} \Psi_{ik}(t) = 0$, including the internal Hamiltonian $\hat{H}_0$ and the system–laser interaction via the dipole moment operator $\tilde{\mu}$), $\alpha(t)$ is a time-dependent penalty factor for the laserfield intensity, and $T$ is the total pulse duration. The individual targets $\tau_k \equiv \langle \Psi_{ik}(t = T) | \Phi_{ik} \rangle$, with desired target states $\Phi_{ik}$, are defined by the $N = 2^n$ corresponding state to state transitions induced by the quantum gate unitary transformation in an $n$-qubit standard basis. In an iterative procedure, derived from the maximization of equation (2) (as outlined in \cite{14}), global population transfer between states of the standard basis or superpositions of those can be optimized.

The definition of the aim of the functional $\sum_{k=1}^{N} |\tau_k|^2$ in equation (2) is insensitive to any phase acquired by the $\tau_k$ during each of the transitions. It has been demonstrated that phase gates can be calculated using transitions between selected superposition states \cite{11}. Phase correctness can be assured analogously \cite{14}. Alternatively, different definitions of the aim in functional (2) for directly optimizing a phase correct unitary transformation have been put forth by Palao and Kosloff \cite{19}.

While phase control can be achieved applying optimal control variants, the time development of phase correctly prepared superposition states (with defined relative phases between the standard basis states) is equally important. These superposition states are produced during most quantum algorithms and processed by subsequent operations, with results dependent on the relative phases. Most prominent are the states of the so-called ‘Hadamard basis’, which in a two qubit system consists of:

$$\frac{1}{\sqrt{2}} (|00\rangle + |01\rangle + |10\rangle + |11\rangle) \quad \frac{1}{\sqrt{2}} (|00\rangle - |01\rangle - |10\rangle + |11\rangle)$$

$$\frac{1}{\sqrt{2}} (|00\rangle + |01\rangle - |10\rangle - |11\rangle) \quad \frac{1}{\sqrt{2}} (|00\rangle - |01\rangle + |10\rangle - |11\rangle).$$

An analysis of the ‘lifetimes’ and the time development of such superposition states during free evolution (i.e. governed by the internal molecular Hamiltonian $\hat{H}_0$) follows in section 3. It will be shown that the relative phase development in multi-qubit superposition states encoded in molecular vibrations displays an additional complexity compared to other implementations.

3. Phase development of vibrational qubit states

We start with the phase development of the standard qubit basis states. The phase development of each $n$-qubit basis state during free evolution $\hat{U}(t - t_0)$ is governed by the frequency $\omega_k$ corresponding to the eigenenergy of the vibrational eigenstate. Thus, the time development for
a superposition state \( \Psi(t_0) = \sum_{k=1}^{2^n} c_k |(q_1 q_2 \cdots q_n)_k \rangle \) of \( n \) qubits is generally defined by:
\[
\hat{U}(t - t_0)\Psi(t_0) = \sum_{k=1}^{2^n} |c_k| e^{i(\theta_k - \omega_k(t - t_0))} |\Psi_k\rangle.
\] (3)

Here, \( c_k \) are the complex coefficients of the standard qubit basis states in the superposition with initial phases \( \theta_k \).

As discussed in section 1, the eigenenergies of molecular vibrational eigenstates which constitute the qubit basis differ. Thus, the free evolution imprints an additional phase \(-\omega_k(t - t_0)\) on each qubit standard basis state, which is of fundamental relevance, since this leads to a change in the relative phases between all the standard basis states in a coherent superposition. This evolution of the relative phases in an \( n \)-qubit superposition state \( \Psi(t_0) \) can be visualized in general by its autocorrelation function:
\[
|\langle \Psi(t_0) | \hat{U}(t - t_0) | \Psi(t_0) \rangle|^2 = \sum_{k=1}^{2^n} |c_k|^4 + \sum_{k=1}^{2^n} \sum_{l>k}^{2^n} 2|c_k|^2|c_l|^2 \cos(\Delta \omega_{kl}(t - t_0)).
\] (4)

As already observed for a two qubit system in [14] any arbitrary absolute phase \( \phi \) of the superposition state, like \( e^{i\phi}|\Psi(t_0)\rangle \), acquired during the time evolution has no experimentally observable effects and consequently does not influence the successful implementation of subsequent quantum gates. This absolute phase will be disregarded in the following. The relevant physical quantity is \( |\langle \Psi(t_0) | \hat{U}(t - t_0) | \Psi(t_0) \rangle|^2 \).

A system of \( n \) qubits spans a basis with a dimension of \( 2^n \), and for vibrational molecular quantum computing, \( \binom{2^n}{2} = 2^n - 1 \) energy spacings \( \Delta \omega_{kl} = \omega_k - \omega_l \) contribute to the development of the relative phases in a superposition state and thus to the autocorrelation (equation (4)). These energy spacings are determined by the different excitation frequencies of the qubit normal modes and the additional energy shift of multiple excited states due to the coupling or entanglement between the normal modes. The oscillation pattern of an autocorrelation reflects the variety of frequency differences \( \Delta \omega_{kl} \), their interferences and the corresponding development of the relative phases. In figure 1, we show the development of the autocorrelation (in black) in several \( n \)-qubit model systems \((n = 2, 4, 6)\) with different molecular parameters, representative for typical IR absorbers.

The short time phase development is dominated by the highest excitation energies of the normal modes and occurs within femtoseconds (figure 1, left panels in black). For an experimental realization of molecular quantum computing, this poses very high demands on the laser pulse synchronization. In addition, the very fast phase development renders simultaneous population and phase optimization much more complex compared to the exclusive optimization of global population transfer. The long time development is determined by small deviations of the energy differences (figure 1, right panels), generated by the coupling between the qubit normal modes. The related timescale usually is in the range of a few picoseconds (figure 1, right panels).

The time for 100% recurrence is proportional to the smallest common multiplicative of the inverse of all energy differences. For the two and four qubit systems presented, there are intermediate recurrences with very high fidelities, arbitrarily close to 100% that emerge within reasonable time intervals. For the six qubit system, however, the fidelity of the recurrence around 4170 fs already decreases to about 90%. Molecular parameters could be adapted by chemical engineering to yield high recurrences in the low picosecond regime, where effects such as population relaxation or other decoherence processes do not play a role (see e.g. four-qubit model system in figure 1 with transition energies of multiples of 600 cm\(^{-1}\)).
Figure 1. Short time (a)–(c) and long time (d)–(f) phase development of superposition states in a two-qubit (2QS), a four-qubit (4QS) and a six-qubit model system (6QS). Displayed are the autocorrelation (black) and the correlation (grey). The correlation refers to a state which is phase correct for a subsequent operation on the second qubit. The transition energies of the qubit normal modes are given in cm$^{-1}$ in the right panels (e)–(f), that of the second qubit is marked in grey, and the coupling between two normal modes is always 8 cm$^{-1}$.

As a general trend, the simulations show that with an increasing qubit basis size, the dephasing generally becomes faster and more pronounced, and the time for sufficiently high recurrences is significantly prolonged. However, the discussed phase evolution can be interpreted as phase gates, which is demonstrated in the following. Based on this new concept, we introduce a possible way to overcome the dephasing problem.

4. New approach to quantum logic operations

As demonstrated, a rapid and individual relative phase development of qubit basis states emerges naturally when molecular vibrational eigenstates are selected to encode qubits. Our idea is to take advantage of this phase development of vibrational superposition states. The free phase evolution during a defined time $T$ can be interpreted as a phase gate and could be utilized to simplify the phase control task during universal quantum logic operations. In a different context, free phase evolution has successfully been applied in non-universal approaches to simulate quantum logic with specially prepared rovibrational superposition states [24, 25]. Here, we will introduce a
concept for the implementation of phase gates on vibrational qubits, as a flexible alternative to laser control, for universal quantum computing.

The general, analytical formula for simulating the development of the fidelity of a desired phase gate $\hat{U}_{\phi_k}$ during free evolution, with target relative phases $\Delta \phi_{kl}$, starting from an initial superposition state $|\Psi(t_0)\rangle$ with initial relative phases $\Delta \theta_{kl}$, is given by:

$$|\langle \Psi(t_0)|\hat{U}_\phi^\dagger \hat{U}(\Delta t)|\Psi(t_0)\rangle|^2 = \sum_{k=1}^{2^n} |c_k|^4 + \sum_{k=1, l>k}^{2^n} 2|c_k|^2|c_l|^2 \cos(\Delta \omega_{kl}\Delta t + \Delta \phi_{kl} - \Delta \theta_{kl}).$$  (5)

The optimal duration $\Delta t$ for $\hat{U}_{\phi_k}$ is obtained when $|\langle \Psi(t_0)|\hat{U}_\phi^\dagger \hat{U}(\Delta t)|\Psi(t_0)\rangle|^2$ is one. The suitability of a qubit system for the implementation of phase gates with high fidelity (arbitrarily close to one) depends on the molecular parameters that determine the $\Delta \omega_{kl}$: the normal mode excitation frequencies and the vibrational coupling parameters, expressed as the shift of the excitation energy for each qubit depending on the actual values of all other qubits. A necessary condition for implementing arbitrary phase gates is that all $2^n$ eigenfrequencies are different. A second requirement can be derived from the fact that for all $\Delta \omega_{kl}$ the condition

$$\Delta \omega_{kl}\Delta t + \Delta \phi_{kl} = n_{kl}2\pi,$$  (6)

needs to be fulfilled to maximize $|\langle \Psi(t_0)|\hat{U}_\phi^\dagger \hat{U}(\Delta t)|\Psi(t_0)\rangle|^2$ (for this purpose the initial phases $\Delta \theta_{kl}$ can be set to zero without loss of generality). Focusing on two selected energy differences $\Delta \omega_{kl}$ and $\Delta \omega_{k'l'}$, the following relation can be set up for $\Delta t$:

$$\Delta t = \frac{(n_{kl} - n_{k'l'})2\pi - (\Delta \phi_{kl} - \Delta \phi_{k'l'})}{\Delta \omega_{kl} - \Delta \omega_{k'l'}.}$$  (7)

All $\Delta \omega_{kl}$ must be different for this equation to be solvable (for $\lim |\Delta \omega_{kl} - \Delta \omega_{k'l'}| \rightarrow 0$, $\Delta t \rightarrow \infty$). Both requirements, different $\omega_k$ and different $\Delta \omega_{kl}$, are primarily encountered in qubit systems encoded in nondegenerate molecular vibrations. For the qubit system parameters we tested, gate durations in the range of a few picoseconds and gate fidelities above 99% were derived.

Based on these results, we propose that quantum logic operations on molecular vibrational qubits can be partitioned into global population transfer, induced by specially shaped ultrashort laser pulses, and successive phase gates through free evolution during a delay between the laser pulses. The two tasks of population control and phase control that involve different timescales are disentangled, which generally reduces the complexity of the total control task. Laserfields that induce the global population transfer desired for a quantum logic operation without restrictions on the phase in general are much easier to optimize. Also, from the duration of the laser-driven switching process plus the free phase rotation, an optimal duration $T$ for the complete, phase correct quantum gate can be derived. Moreover, a reduction of the system–laser interaction is achievable.

Exemplarily, we present two applications of this concept in the two qubit model system of figures 1(a) and (d), with parameters given in table 1. The molecular parameters are the excitation frequencies of the two qubit normal modes $\omega_1 = 2000$ cm$^{-1}$ and $\omega_2 = 1400$ cm$^{-1}$ (spanning the frequency region of many IR-active groups as e.g. carbonyl modes, amide modes or CH-deformation modes) and the coupling parameter which introduces a negative energy shift of 8 cm$^{-1}$ to the $|11\rangle$ state (from 3400 to 3392 cm$^{-1}$).
Table 1. Parameters for the simulations of free evolution phase gates in the model system of figure 1(a). $\Delta \omega_{kl}$ are the frequency differences in the qubit basis, $\Delta \theta_{kl}$ are the initial relative phases and $\Delta \phi_{kl}$ are the desired target relative phases.

| $l - k$ | $\Delta \omega_{kl}$(cm$^{-1}$) | $\Delta \theta_{kl}$ | $\Delta \phi_{kl}$ | $\Delta \theta_{kl}$ | $\Delta \phi_{kl}$ |
|----------|-----------------|-----------------|-----------------|-----------------|-----------------|
| |                  | $\Delta \theta_{kl}$ | $\Delta \phi_{kl}$ | $\Delta \theta_{kl}$ | $\Delta \phi_{kl}$ |
| 00 – 01  | 1400            | -0.13\pi         | 0               | 0               | 0               |
| 00 – 10  | 2000            | -0.79\pi         | 0               | 0               | -               |
| 01 – 11  | 1992            | 0.05\pi          | 0               | 0.5\pi          | -               |
| 10 – 11  | 1392            | 0.71\pi          | 0               | 0.5\pi          | 0               |
| 00 – 11  | 3392            | -0.08\pi         | 0               | 0.5\pi          | -               |
| 01 – 10  | 600             | -0.66\pi         | 0               | 0               | -               |

4.1. Phase adjustment of a qubit flip gate

In a first example we apply the concept of free evolution phase gates to ensure the phase correct implementation of a CNOT gate switching the second qubit (CNOT$_2$).

The corresponding CNOT$_2$ laser field has been optimized for global population transfer (> 99%) with the standard algorithm (derived from equation (2)). As already mentioned, this yields faster and simpler results for most target times $T$ than any algorithm including conditions on the phase. In figure 2(a) the CNOT$_2$ gate laser field is shown in grey (top panel), together with its fidelity development (bottom panel). The fidelity is defined according to [19, 23] as

$$\frac{\left|\sum_k^{N} \tau_k\right|^2}{N^2}.$$  (8)

At the end of the CNOT$_2$ laser pulse, at 3000 fs, a fidelity of only $\approx 38\%$ is reached despite correctly driven population transfer, due to the arbitrary development of the relative phases of the $\tau_k$. The complex coefficients $\tau_k = \langle \Psi_k(T) | \Phi_{f_k} \rangle$ (projections of the propagated states on to the target states) at the end of the laser interaction are displayed in figure 2(b), also in grey. The correct population transfer is reflected in the unit length of the arrows. The phases which are acquired during each state-to-state transition are not equal, as required for a phase correct CNOT operation. The laser pulse therefore really induces a CNOT$_2$ gate plus a phase gate.

During a free evolution of the quantum system subsequent to the CNOT$_2$ laser pulse, with a specific duration derived from the simulation with equation (5) and parameters given in table 1, the phases of all transitions are synchronized (see figure 2(b), black arrows). At this point, the complete quantum gate is phase correct and the fidelity rises to virtually 100% in figure 2(a) (bottom panel, displayed in black). This simple scheme can be straightforwardly applied to ensure the phase correctness of all qubit flip gates (e.g. CNOT, NOT and Hadamard) with laser fields optimized for global population transfer.
4.2. Conditional phase gate and the quantum Fourier transform (QFT)

Pure phase gates can be realized solely by free evolution. As an example, we implement the conditional quantum phase gate

\[ U_{\ket{11}} = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & e^{i\pi/2} \end{pmatrix}, \]  

again in the two-qubit model system with parameters given in figure 1 and table 1. This gate is part of a two-qubit QFT constructed from essential quantum gates in a sequence adapted from [26]

\[ \text{QFT}_4 = \text{CNOT}_2 \text{CNOT}_1 \text{CNOT}_2 H_2 U_{\ket{11}} H_1, \]
where $H_i$ are the Hadamard gates on the qubit $i$ and the sequence is applied in a time order from right to left.

The free evolution time of 1000.7(±0.6) fs needed for the realization of $U_{i\{11\}}$ with a fidelity of higher than 99% is extracted from the simulation according to equation (5) and parameters given in table 1. The development of the fidelity of $U_{i\{11\}}$ is displayed in figure 3 in black.

The laser pulse sequence we optimized for the total QFT$_4$ is shown in figure 4 (upper panel). The Hadamard gates $H_i$ applied in this sequence are implemented phase correctly already by the system–laser interaction. The CNOT$_i$ gates are realized with our new scheme, by decomposition into laser-driven global population transfer and free evolution phase gates. Figure 4 (lower panel) displays the population development induced by the laser pulse and delay sequence exemplarily with $|01\rangle$ as initial state. During the periods exhibiting no population transfer, the phases are adjusted correctly by free evolution. For the implementation of the total QFT$_4$-transformation, a fidelity of over 99% is accomplished.

4.3. Robust implementation of global and phase correct quantum gate sequences

We have shown that the phase evolution occurring in vibrational molecular quantum computing is not separable for the individual qubits. This feature can be used to implement phase gates and decompose a quantum gate into laser-driven population control and subsequent phase synchronization by free evolution. Yet the challenge still exists that the phase evolution and thus the phase control becomes very complex with an increasing number of qubits.

Nevertheless, single qubits can be selectively addressed and single qubit transitions are globally driven by specially shaped laserfields. Thus, a more robust and simple application of the free phase evolution is attained, when only the basis states which are coupled by the next quantum gate laserfield are phase adjusted. For example, a switching laserfield addressing the last of $n$ qubits, couples the states $|(q_1q_2q_3\cdots 0)_m\rangle$ and $|(q_1q_2q_3\cdots 1)_m\rangle$ for all $m$ given by

**Figure 3.** The black graph shows the time development of the fidelity for the $\hat{U}_{i\{11\}}$ phase-gate. The long time development is displayed in the left panel and a zoom of the region around the total time $T$ of $\hat{U}_{i\{11\}}$, marked with the black circle in the right panel. The grey graph describes the adjustment of the right relative phase between $|00\rangle$ and $|01\rangle$ concurrently with the right relative phase between $|10\rangle$ and $|11\rangle$, which ensures the correct action of a subsequent quantum gate on the second qubit. The grey circle marks the delay time chosen for $U_{i2}$ in the alternative QFT sequence.
the possible values of the other qubits. For the third qubit, transitions like $|q_1q_20\cdots q_m\rangle \leftrightarrow |q_1q_21\cdots q_m\rangle$ are selectively addressed, and so on. Therefore, the only phases relevant are those between the respective pairs of standard basis states—$2^{n-1}$ in contrast to $(2^n-1)$ when all relative phases in a superposition state of an $n$-qubit basis have to be synchronized.

For all model systems discussed in section 3, figure 1 also displays the correlation functions of superposition states, solely adjusting the phases between basis states that are coupled during a subsequent switching process of the second qubit (in grey). The value reached for the correlation function is always greater than or equal to that of the autocorrelation function and recurrences of high fidelity are more abundant. Also, the phase relations decay more slowly, since the $2^{n-1}$ frequency differences $\Delta\omega_{kl}$ defining the fast evolution are equal to the excitation energy of the addressed qubit. This excitation energy spans a small frequency range with a width determined by the prevalent coupling between the qubit normal modes. Consequently, the complexity of the short time phase development in multi-qubit superposition states is reduced to that of a virtual one qubit system, and also the complexity of the long time development is significantly reduced. Thus, the relevant phase evolution here is to a good approximation independent of the size of the qubit system and renders this approach more scalable.

Applying this more robust scheme in the QFT$_4$ presented in the previous paragraph, the phase gate $U_{|11\rangle}$ can be alternatively implemented adjusting only the relative phases between the basis states $|00\rangle$ and $|01\rangle$ as well as $|10\rangle$ and $|11\rangle$. The fidelity of this operation labelled $U_{i2}$ is displayed for comparison with the original $U_{|11\rangle}$ in figure 3, in grey. The grey circle marks the delay time chosen for this alternative conditional rotation gate, where the fidelity of the unitary transformation $U_{|11\rangle}$ itself is close to 0%.

Let us consider the action of the alternative QFT$_4$ sequence applying this robust scheme, exemplarily on the qubit basis state $|01\rangle$ (see also table 2). The $H_2$ gate following $U_{i2}$ in the alternative QFT$_4$ sequence then acts on the superposition state $\frac{1}{\sqrt{2}}(|01\rangle + e^{i\phi}|11\rangle)$ with an
Table 2. Action of the alternative QFT$_4$ sequence on the qubit basis state $|01\rangle$. $\phi$ and $\phi_m$ denote the ‘arbitrary’ phases generated by the respective switching laserfield. They are the same for each transition in the qubit basis, i.e. irrespective of the initial qubit basis state. When read without the arbitrary phases $\phi$ and $\phi_m$, the table shows the development governed by a sequence of laserfields for phase correct quantum gates. Any global phases are disregarded.

| Start | Laserfield/delay | Resulting state |
|-------|-----------------|-----------------|
| $|01\rangle$ | $H_1$ | $\frac{1}{\sqrt{2}}(|01\rangle + |11\rangle)$ |
| | $U_{12}$ | $\frac{1}{\sqrt{2}}(|01\rangle + e^{i\phi}|11\rangle)$ |
| | $H_2$ | $\frac{1}{\sqrt{2}}((|00\rangle - |01\rangle) + e^{i\phi} (i|10\rangle - i|11\rangle))$ |
| | CNOT$_2$ | $\frac{1}{2}((e^{i\phi} |00\rangle - e^{i\phi} |01\rangle - e^{i\phi} i|10\rangle + e^{i\phi} i|11\rangle)$ |
| | $D_1$ | $\frac{1}{2}((|00\rangle - i|10\rangle) + e^{i\phi} (-|01\rangle + i|11\rangle))$ |
| | CNOT$_1$ | $\frac{1}{2}((e^{i\phi} |00\rangle + e^{i\phi} i|01\rangle - e^{i\phi} i|10\rangle - e^{i\phi} i|11\rangle)$ |
| | $CNOT_2$ | $\frac{1}{2}((|00\rangle + i|01\rangle - |10\rangle - i|11\rangle)$ |
| | $D_{12}$ | $\frac{1}{2}((|00\rangle + i|01\rangle - |10\rangle - i|11\rangle)$ |

arbitrary relative phase $e^{i\phi}$. The subsequent CNOT$_2$ laserfield can be applied as hitherto. Before the next CNOT$_1$ laser pulse, the relative phases $|00\rangle \pm i|10\rangle$ as well as $|01\rangle \pm i|11\rangle$ have to be set during an extra time delay $D_1$, now disregarding any relative phases between states with different value of qubit two. The next two steps of a time delay $D_2$ and a CNOT$_2$ laserfield follow the same procedure. The robust QFT$_4$ sequence is completed by a final delay $D_{12}$, during which the phases of all qubits are correlated in the right way. The arbitrary relative phases $e^{i\phi}$ and $e^{i\phi_m}$, produced by the switching laserfields and during the delays are the same for each transition starting from standard basis states. As a consequence, this alternative QFT$_4$ sequence as a whole is phase correct and independent of the basis (standard or Hadamard) it acts on. With this approach of only adjusting the relative phases of selected pairs of qubit states during the whole computational process, the QFT$_4$ fidelity is still above 99%, while the total duration stays approximately the same.

5. Effects of the CEP phase of the driving laserfield

In earlier investigations, we found that the global population transfer induced by specially shaped laserfields acting as quantum gates is independent of the absolute phase of the incident laserfield. We define the absolute phase at the intensity maximum of the laserfield in analogy to the CEP and in the following refer to it as ‘CEP’. The population transfer is dependent only on the relative phases in sub pulse sequences (see for example [15]). Here, we show that the phase correctness of a quantum gate is, however, directly linked to the CEP of the driving femtosecond laserfield. Consider for example a CNOT$_2$ laserfield in the two qubit model system of table 1 with a CEP shifted by $\phi_{\Delta\text{CEP}}$ with respect to the optimized one. We found, that it performs a unitary transformation with additionally induced relative phases $\phi_{\Delta\text{CEP}} \neq 0$ to the laser-driven
Figure 5. (a) Action of a CNOT$_2$-laserfield, optimized phase correctly, with different CEPs on a two qubit maximum superposition state. Displayed are the vector representations of the complex coefficients of the standard basis states in the superposition, in the initial or input state to the very left, and in the final or output states depending on the $\phi_{\Delta\text{CEP}}$ given in $n\pi$ of the CNOT$_2$-laserfield with respect to the originally optimized field (0$\pi$). (b) Measurement probabilities for $|01\rangle$ and $|11\rangle$ from the final state of the H$_1$-H$_2$-CNOT$_2$-H$_2$-H$_1$-sequence with different CEPs of the CNOT$_2$ laserfield (again with respect to the $\phi_{\Delta\text{CEP}}$ relative to the optimized one). The initial state was $|01\rangle$ and the correct final state should be $|11\rangle$. For $\phi_{\Delta\text{CEP}} = \pi$ the probability for measuring $|11\rangle$ is 0%, as the quantum system is in state $|01\rangle$ with 100% probability.

states $|10\rangle$ and $|11\rangle$, e.g. in the Hadamard basis

$$\frac{1}{\sqrt{2}}(|00\rangle - |01\rangle + |10\rangle - |11\rangle) \rightarrow \frac{1}{\sqrt{2}}(|00\rangle - |01\rangle - e^{i\phi_{\Delta\text{CEP}}} |10\rangle + e^{-i\phi_{\Delta\text{CEP}}} |11\rangle)$$

(neglecting any acquired global phase). For $|00\rangle$ and $|01\rangle$ the phase evolution is independent of the CEP of the driving laserfield. This is shown in figure 5(a). For $\phi_{\Delta\text{CEP}} = 0$, the states $|10\rangle$ and $|11\rangle$ are interchanged, switching their phases in the superposition as desired. For $\phi_{\Delta\text{CEP}} \neq 0$, the laserfield does induce the population transfer $|10\rangle \leftrightarrow |11\rangle$, but not the phase correct transition and the relative phases between $|10\rangle$ and $|11\rangle$, as well as their phases relative to $|00\rangle$ and $|01\rangle$ are set incorrectly. For shifted CEPs, the relative phases in the resulting superposition state can be readjusted by an extra time delay according to the scheme presented in the previous section. This applies to all CNOT and NOT (qubit flip) gates.

A special case of the CEP dependence arises for the Hadamard gate. For example, a H$_2$ laserfield with a shifted CEP will drive the right population transfer with high fidelity, e.g.

$$|X0\rangle \rightarrow \frac{1}{\sqrt{2}}(|X0\rangle + e^{-i\phi_{\Delta\text{CEP}}} |X1\rangle), \quad |X1\rangle \rightarrow \frac{1}{\sqrt{2}}(e^{i\phi_{\Delta\text{CEP}}} |X0\rangle - |X1\rangle),$$

where $X$ is the arbitrary state of the passive qubit (again, disregarding any absolute phase). The relative phases generated in the superpositions depend on the CEP shift of the laserfield. A CEP-shifted H$_2$ laserfield is still self-inverse, as the phases $\phi_{\Delta\text{CEP}}$ cancel when it is applied twice. However, the reverse transformation from the correct superposition to the standard basis
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states $\frac{1}{\sqrt{2}}(|X0⟩ + |X1⟩) \rightarrow |X0⟩$ and $\frac{1}{\sqrt{2}}(|X0⟩ − |X1⟩) \rightarrow |X1⟩$ cannot be accomplished by a CEP-shifted laserfield due to the additional relative phase $\phi_{\text{CEP}}$ that is accumulated during the transitions in (11).

We want to emphasize that quantum gate sequences can be viewed as a type of molecular wavepacket interferometry experiment in which the outcome reflects variations of the CEP of the driving laserfield(s). As a first experiment, we propose the sequence $H_1 - H_2 - \text{CNOT}_2 - H_2 - H_1$ starting from $|01⟩$, for which the phase correct quantum gate sequence gives the result $|11⟩$. With stabilized CEPs of the $H_i$ laserfields, a variation of the CEP of the CNOT$_2$ laserfield alone will generate different results for the subsequent, final Hadamard gates. As a consequence, the projection into the standard basis by the final Hadamard gates yield $|11⟩$ with a different probability for each $\phi_{\text{CEP}}$ of the CNOT$_2$ laserfield, as shown in figure 5(b). The probability for measuring the input state $|01⟩$ rises as the probability for measuring $|11⟩$ falls. For $\phi_{\text{CEP}} = \pi$, the intermediate superposition state produced by the CNOT$_2$ gate is projected on to the initial $|01⟩$ state with virtually 100% probability (see also figure 5(a)).

In summary, the correct implementation of quantum gate sequences and consequently their results depend sensitively not only on the timing of subsequent femtosecond to picosecond laser pulses but also on their phase, requiring a well-stabilized CEP. Both parameters are closely interconnected with the relative phase development in the qubit basis through the coherence transferred from the laserfields to the excited vibrational eigenstates.

6. Conclusions

In this paper, we have simulated the relative phase development in superposition states which emerges during a quantum computation process in a qubit basis encoded in molecular vibrational eigenstates. We have demonstrated how this free phase evolution can be interpreted as phase gates and can in principle be utilized to correct undesired phases acquired during system–laser interaction and to implement arbitrary phase gates. Thus, we suggest to decompose quantum gate sequences into laser-driven qubit switching processes followed by a phase adjustment during free evolution, corresponding to defined delays between subsequent laserfields. Due to the fast (femtosecond to sub-femtosecond) decay time of the desired relative phases in multi-qubit superposition states, we find that subsequent quantum gate laserfields in a corresponding experiment need to be synchronized very well. This poses an experimental challenge which becomes more demanding with increasing dimension of the qubit systems. For alleviation, we propose a way to virtually reduce the phase space considered to that of the qubit addressed during the next logic operation only. This way, robust and overall phase correct quantum gate sequences as well as complete algorithms can be implemented more flexibly, according to experimental conditions.

In addition, we showed that the absolute phase or CEP of the driving laserfield is also crucial for the phase correct implementation of quantum gate laserfields and algorithms in molecular quantum computing with vibrational eigenstates. The phase coherence, which is built up, while the first quantum gate laserfield is acting on a multi-qubit basis has to be carefully conserved throughout the whole algorithm to yield a correct result. For this to be accomplished, the rapid relative phase development in the vibrational multi-qubit system has to be taken into account and timing and phases of the switching laserfields have to be precisely controlled and stabilized. Finally, we have proposed a sequence of quantum gates including a CNOT-gate in a two-qubit
basis for which the measurement outcome will be sensitive to the variation of the CEP of the CNOT laserfield.

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