Quantum Logic Approach to Wave Packet Control

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We study control of wave packets with a finite accuracy, approaching it as quantum information processing. For a given control resolution, we define the analogs of several quantum bits within the shape of a single wave packet. These bits are based on wave packet symmetries. Analogs of one- and two-bit gates can be implemented using only free wave packet evolution and coordinate-dependent ac Stark shifts applied at the moments of fractional revivals. As in quantum computation, the gates form a logarithmically small set of basis operations which can be used to approximate any unitary transformation desired for quantum control of the wave packet dynamics. Numerical examples show the application of this approach to control vibrational wave packet revivals.

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Advances in femtosecond control of coherent molecular dynamics have stimulated theoretical [1] and experimental [2] efforts aimed at realizing logical gates and quantum computing algorithms in atoms and molecules, especially using the feedback control methods [3,4]. In this Letter we pass the connection between quantum control and quantum computation in the opposite direction, studying how the perspective of quantum information processing can help to implement goals of quantum control—specifically, the control of wave packet dynamics.

An N-level system can carry only a $K \sim \log_2 N$ amount of information, just like a K-bit quantum computer. This suggests that the complexity of controlling an N-level system might scale similarly, i.e., $\sim \log_2 N$. In a K-bit quantum computer, any unitary transformation within the $N = 2^K$-dimensional Hilbert space can be approximated using $O(K = \log_2 N)$ basic one- and two-bit gates. We show how one can identify and implement $O(\log_2 N)$ basic operations on a wave packet, sufficient to approximate any unitary evolution with the resolution of $1/N$th of the orbit.

Questioning the number of basic elementary operations needed for control refers to the question of what resources—laser pulse bandwidth, peak intensity, number of pixels in the pulse shaper, etc.—are required. There is a tradeoff between the complexity of the control pulse and the control efficiency. On the one hand, if a laser pulse can be shaped on an arbitrarily fine time scale, one can almost always generate N^2 different parameters needed to implement any unitary transformation in an N-level system in a finite time [5,6]. The number of experimental "knobs" (e.g., the flexibility of the pulse shaper) can be decreased at the expense of time needed and accuracy of the control. Even with a fixed (duration and intensity) pulse one should be able to approximate any desired transformation by applying the same pulse many times at the expense of exponentially growing control time [5]. These, however, are statements about the *existence* of a solution and rely on the assumption that N is fixed.

Most *constructive* solutions rely on either intuitively clear schemes for specific state-to-state transformations (most notably Brumer-Shapiro and Tannor-Kosloff-Rice approaches [4]) or on the numerical optimization procedure. There is a very limited number of general constructive schemes, all requiring $O(N)$ different resonant frequencies: for state-to-state transformation, see an elegant solution in [7], and for generating the desired $N \times N$ matrix in $O(N^2)$ steps, see, e.g., [8].

Analogs of logical gates would offer algorithmic and physical prescriptions of how the control of both states and matrices can be implemented with a sequence of standard transformations. Then, one may be able to approach the speed of control associated with a quantum computer using a small ($\sim \log_2 N$) number of basic elementary operations.

To implement a quantum logic approach to control, we take a time-domain (wave packet) perspective. In the wave packet experiments the number of levels involved may change during control operations; controlling a wave packet means controlling its shape and position. We define analogs of logical qubits based on the wave packet shape and symmetries, independently of the number of levels in the wave packet. We then construct analogs of logical gates for systems with quadratic dispersion, using molecular vibrations as an example. The gates utilize only free wave packet evolution and phase kicks made by a relatively weak coordinate-dependent ac Stark shift, applied at the moments of fractional wave packet revivals. We show that free evolution in combination with phase kicks implements a complete set of logical gates for a wave packet. Hence, it allows for complete control of the wave packet evolution within the accuracy defined by the number of qubits.

Consider a wave packet $\Psi(t) = \sum_n C_n(t)\phi_n$, with amplitudes $C_n(t) \propto \exp(-iE_n t)$ distributed around a state

$|n_0\rangle$ (E_n, ϕ_n are the energies and wave functions of the states). Just as with a superposition of optical waves, the wave packet can be described by a carrier and an envelope [9]. The carrier is given by the stationary wave function ϕ_{n_0} while the wave packet envelope f is defined as

$$f(t, \tilde{\theta}) \equiv \sum_n C_n(t) \exp[i(n - n_0)\tilde{\theta}]. \tag{1}$$

It moves along the classical orbit associated with $|n_0\rangle$, and $\tilde{\theta}$ is the phase (angle) of that motion [9]. For equidistant spectrum $E_n - E_{n_0} = \omega_0(n - n_0)$ the envelope is $f(t, \tilde{\theta}) = f(\tilde{\theta} - \omega_0 t) = f(\theta)$, and its shape is unchanged during the motion, which occurs with the period $T_0 = 2\pi/\omega_0$. Anharmonicity in the spectrum $E_n - E_{n_0} = \omega_0(n - n_0) + \Omega(n - n_0)^2$ leads to the wave packet spreading and revivals: after time $T_{\rm rev} = 2\pi/\Omega$ the wave packet relocalizes (revives) at its initial position. At the fractional revival time $T_{\rm rev}/2M$ the wave packet splits into M *orthogonal* clones $F_i(\theta)$ spaced by $\Delta\theta = 2\pi/M$ [10].

Figure 1 is obtained by propagating the initial wave packet on the Cl_2-like potential modeled as a Morse oscillator $V(x) = D[1 - e^{-ax}]^2 - D$ with $x = R - R_{\rm eq}$, $D = 2.47$ eV, and $a = 1.07$ a.u. The wave packet with reduced mass $m = 31\,270$ a.u. starts as Gaussian, $C_v(t = 0) \propto e^{-(v-v_0)^2/\sigma^2}$, with $v_0 = 38$ and $\sigma = 4/\sqrt{\ln 2}$ (i.e., FWHM = 8 levels). Near $v = 38$ the vibrational period is $T_0 = 130$ fs and $T_{\rm rev} \approx 8.31$ ps. The fractional revival about $t = T_{\rm rev}/8$ is immediately visible in the envelope representation [Eq. (1) and Fig. 1, inset], which removes carrier oscillations and brings out the underlying symmetries in the wave function.

The definition of wave packet "bits" in terms of $f(\theta)$ is illustrated in Fig. 2 for a two-bit system. These states are defined as four orthogonal superpositions of the "basis" wave packets F_i obtained from the initial wave packet at the fractional revival time $T_{\rm rev}/8$ (the four orthogonal clones in Fig. 1, inset). In general, for K bits we divide $\tilde{\theta} = 0 \ldots 2\pi$ into 2^K equal intervals. The states of the bits are determined by the *symmetry* of $f(\theta)$ with the resolution increasing in powers of 2. Reminiscent of the fast Fourier transform, the lowest bit distinguishes between $f(\theta)$ even or odd on the whole orbit, the second bit distinguishes parity on every half of the orbit, the third would do it for every quarter, etc. For K-bit computation, structures finer than $2\pi/2^K$ in $f(\theta)$ are unresolved.

Thus, all basis wave packets in phase correspond to the ground state $|0, \ldots, 00\rangle$. A flip of the first (lowest) bit corresponds to multiplying $f(\theta)$ by $e^{i\pi} = -1$ on the second half of the phase orbit, $\theta = \pi \ldots 2\pi$. A flip of the second bit corresponds to multiplying $f(\theta)$ by $e^{i\pi}$ on the second and the fourth quarters of the orbit, thus doubling the modulation frequency. To flip the third bit one doubles the modulation frequency again, etc.

Each wave packet bit distinguishes, on its own scale, not only between "odd" and "even," but also between "left" and "right." As is clear from Fig. 2, the states $(|0\rangle + |1\rangle)/\sqrt{2}$ and $(|0\rangle - |1\rangle)/\sqrt{2}$ of the first bit correspond to the wave function concentrated on the left side ($\theta < \pi$) or on the right side ($\theta > \pi$) of the whole orbit. The states $(|0\rangle + |1\rangle)/\sqrt{2}$ and $(|0\rangle - |1\rangle)/\sqrt{2}$ of the second bit describe the wave function concentrated on the left or the right sides of each half orbit, the third bit would do the same for each quarter orbit, and so on.

We now identify analogs of single-bit and two-bit logical operations and demonstrate them for a two-bit system in a Morse oscillator. We begin with an analog of Rabi oscillation in a single qubit.

As seen from Fig. 2, flipping the first bit requires changing the phases of two basis wave packets on the right side of the orbit by π. This operation flips the first bit independently of the state of the second bit, i.e., performs both $|00\rangle \Leftrightarrow |01\rangle$ and $|10\rangle \Leftrightarrow |11\rangle$ transitions. In general, for the jth bit the phase orbit $\theta = 0 \ldots 2\pi$ is divided into $J = 2^j$ equal parts, and on every second one of these parts the envelope $f(\theta)$ is multiplied by $\exp(i\phi)$. This procedure performs the operation

$$U_R = e^{i\phi/2}\begin{pmatrix} \cos\phi/2 & -i\sin\phi/2 \\ -i\sin\phi/2 & \cos\phi/2 \end{pmatrix} \tag{2}$$

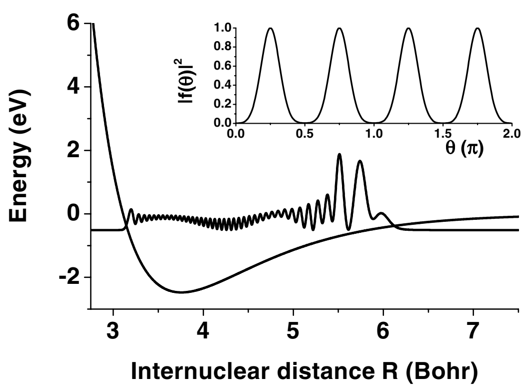

FIG. 1. Wave packet ($|\Psi|^2$) made of ~8 eigenstates in a Morse oscillator at $t = T_0/8 + T_{\rm rev}/8$ and its envelope as the function of the classical angle θ (inset).

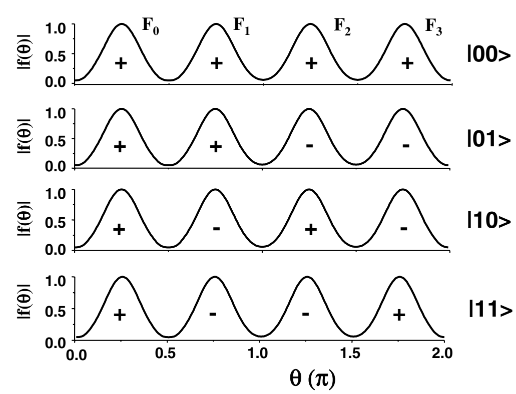

FIG. 2. Four orthogonal basis states of the envelope function for the two-bit system. The signs give relative phases of the basis wave packets.

on the jth bit while all others are unaffected. Equation (2) can be derived by applying the above procedure to the $(|0\rangle \pm |1\rangle)/\sqrt{2}$ states of the jth bit: one is unaffected while the other acquires a global phase $\exp(i\phi)$. Up to the phase factor $e^{i\phi/2}$ U_R is equivalent to Rabi oscillation between the states $|0\rangle$ and $|1\rangle$ of the jth qubit.

A phase shifter can be implemented by a local modification of the potential, i.e., by nonresonantly coupling it to another potential energy surface. This results in an ac Stark shift, i.e., slightly shifts the potential near the turning point while the laser field is on. A wave packet moving through the turning point acquires extra phase $e^{i\Delta S}$ where ΔS is the change in the classical action. The nonresonant pulse must be timed within the window that the wave packet spends near the turning point. This restricts the number of basis wave packets that can be cleanly addressed, the number of qubits that can be encoded, and, hence, the control over the wave packet shape: a one-bit control does not resolve below a half orbit, two-bit control does not resolve below a quarter orbit, etc.

Numerical simulations [Fig. 3(a)] show how such a phase shifter flips the first bit for the same initial wave packet and surface as in Fig. 1 (see the Fig. 3 caption for the pulse parameters). The pulse nonresonantly couples the main surface near the inner turning point to an auxiliary surface also chosen as a Morse oscillator. Shifted down by the energy of one photon, the auxiliary surface is $V'(x) = D'\{1 - \exp[-a'(x + \delta)]\}^2 - D' + \Delta E$ with $D' = 5.44$ eV, $\Delta E = 4.02$ eV, $\delta = 0.2$, and $a' = 1$ a.u. The electronic dipole matrix element is assumed R independent. Figure 3(a) shows that the phase of the last two basis wave packets is flipped by π, as desired. Errors such as the narrowing or widening of the basis wave packets are visible in Fig. 3 but remain unresolved (until accumulated) by the two-bit description, which does not see below a quarter orbit.

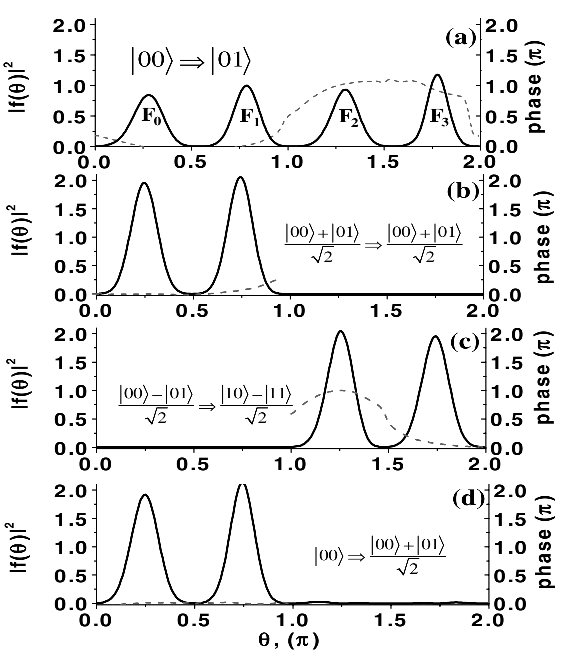

FIG. 3. Numerical simulations of first bit flip (a), CNOT-type gate (b),(c), and Hadamard transform on the first bit (d). Transitions are labeled on the panels. Solid line: $|f(\theta)|^2$; dashed line: phase of $f(\theta)$. Pulse envelope is $e^{-(t/\sigma_T)^8}$. (a) FWHM $= T_0/2 = 65$ fs with coupling $\mu E = 5.47 \times 10^{-3}$ a.u., (b),(c) FWHM $= T_0/4 = 32.5$ fs and $\mu E = 6.04 \times 10^{-3}$ a.u., (d) FWHM $= T_0/2 = 65$ fs and $\mu E = 5.47 \times 10^{-3}/\sqrt{2}$ a.u.

A phase shifter can also be used for multibit gates. For K bits, phase shifting of a single basis wave packet results in a K-bit operation. Figures 3(b) and 3(c) show the result of numerical simulation for the controlled-NOT (CNOT)-type gate in the two-bit system. Applying the laser pulse for the quarter period when the wave packet F_2 passes the turning point does not change the second bit if the first bit is in the state $(|0\rangle + |1\rangle)/\sqrt{2}$ [left, Fig. 3(b)], but flips the second bit if the first one is in $(|0\rangle - |1\rangle)/\sqrt{2}$ [right, Fig. 3(c)].

Changing the phases of basis wave packets is not sufficient — one also needs to change their relative amplitudes. This yields the ability to approximate an arbitrary single-bit gate. For example, the Hadamard states of the first bit used in the previous example require merging half of the basis wave packets with their counterparts which are a half orbit away. To implement the Hadamard transform on the first bit we have to transfer the state $|00\rangle$ into $(|00\rangle + |01\rangle)/\sqrt{2}$ (left), the state $|01\rangle$ into $(|00\rangle - |01\rangle)/\sqrt{2}$ (right), and likewise for the states $|10\rangle$ and $|11\rangle$ — all using the same operation.

Wave packet spreading and revivals give a natural way to accomplish this goal. Indeed, waiting for a fractional revival $T_{\rm rev}/4$ and correcting phases one transfers a single basis wave packet into two packets on the opposite sides of the orbit with an arbitrary relative phase. Similarly, four packets with arbitrary phases can be obtained from a single wave packet at $T_{\rm rev}/8$, using the phase operation. Conversely, any four or two wave packets can be merged into one. This gives a guideline for implementing Hadamard transforms. Figure 3(d) shows a simulation of the revival-based Hadamard transform $\hat{H}_1$ for the first bit, for the same model system as above. It is achieved by applying a $\pi/2$ phase shift on the $\theta > \pi$ part of the orbit and waiting for $T_{\rm rev}/4$. Hadamard transforms for higher bits are less trivial, but still possible [11].

As an example of possible control schemes, Fig. 4 shows how the bit operations control the revival structure and evolution within the vibrational period. The pulse parameters (see Fig. 4) are well within the present-day technology. Figure 4(a) shows $\langle R(t)\rangle = \langle \Psi(t)|R|\Psi(t)\rangle$ of the same vibrational wave packet as used for Figs. 1 and 3. Oscillations of the initially localized wave packet are damped by its spreading. Strong relocalization of the wave packet occurs near the half ($t = T_{\rm rev}/2$) and the full revival.

The only difference between $\Psi(T_{\rm rev}/4)$ and $\Psi(3T_{\rm rev}/4)$ is that the first bit is flipped. Hence, flipping the first bit at

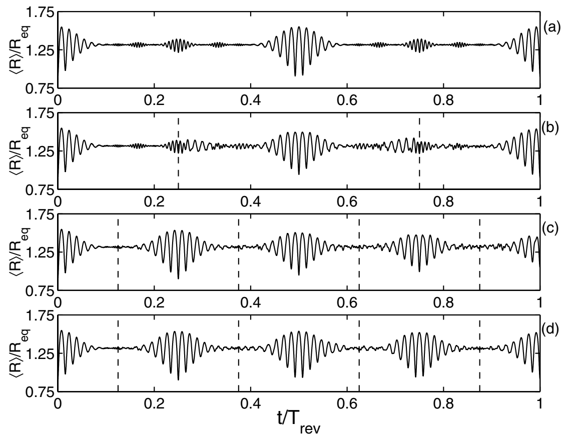

FIG. 4. Control of wave packet revivals by logical operations. Dashed lines show when short control pulses with envelope $e^{-(t/\sigma_T)^8}$ are applied. (a) Free evolution. (b) Each operation uses FWHM $= 65$ fs pulses with $\mu E = 5.47 \times 10^{-3}$ a.u. (c) Each operation uses two FWHM $= 32.5$ fs pulses with $\mu E = 6.04 \times 10^{-3}/\sqrt{2}$ a.u. applied to packets 2 and 4. (d) Each operation uses two FWHM $= 32.5$ fs pulses with $\mu E = 6.04 \times 10^{-3}/\sqrt{2}$ a.u. applied in an alternating manner to packets 1, 3 and 0, 2.

$T_{\rm rev}/4$ will bring the system to $3T_{\rm rev}/4$. This is shown in Fig. 4(b): the revival near $t = T_{\rm rev}/2$ is now π out of phase compared to $t = T_{\rm rev}/2$ but identical to $t = T_{\rm rev}$ in Fig. 4(a). Thus, the arrival of the revived wave packet to the turning point is shifted by half of the vibrational period $T_0/2$. Small oscillations of $\langle R \rangle$ immediately after the first bit-1 flip are the result of small errors of the operation. However, these errors are removed by the second flip of the first bit at $t/T_{\rm rev} = 3/4$.

The difference between $\Psi(T_{\rm rev}/8)$ and $\Psi(7T_{\rm rev}/8)$ is a $\pi/2$ rotation of the second bit. The control pulse in Fig. 4(c) performs a $\pi/2$ rotation of bit 2 at $t/\mathrm{T}_{\rm rev} = 1/8, 3/8, 5/8, \ldots$. Now full revivals are accelerated to occur at every $T_{\rm rev}/4$. The amplitude of subsequent revivals decreases: the errors add constructively. Here every $\pi/2$ rotation was implemented by adding a $\pi/2$ phase shift to packets 1 and 3. Alternatively, one can first apply a $\pi/2$ shift to packets 0 and 4 at $T_{\rm rev}/8$ followed by a $\pi/2$ shift to packets 1 and 2 at $3T_{\rm rev}/8$, etc. (i.e., alternating $\pi/2$ and $-\pi/2$ rotations of the second bit); see Fig. 4(d). Now the errors add destructively: the amplitude no longer decreases as in 4(c).

We have illustrated the ideas of the method using vibrational wave packets, but the same approach can be applied to many other systems with regular spectra, including classical waveguides. A study of the rotational wave packets will be published elsewhere [12]. Note that none of the existing intuitive control schemes is suited for the kind of control shown in Fig. 4. They either require exact knowledge of all the amplitudes C_n (see [7]) or rely on real transitions and absence of spreading (see [4]). A numerical or an experimental optimal control scheme could probably find the same nonresonant mechanism as our solution, but it is well known that such solutions are always complex and hard to interpret.

To summarize, we studied the control of wave packets with a finite resolution—$1/N$th of the phase-space orbit. We have introduced a mechanism of control, which utilizes free wave packet evolution and short kicks by a coordinate-dependent ac Stark shift applied at the moments of fractional revivals. We have shown how, using the envelope representation of the wave packet, one can introduce qubitwise description of the system based on progressive scale symmetries of the wave packet envelope. Analogs of $\sim\log_2 N$ single-bit and two-bit gates can be implemented in a relatively simple manner. Thus, one needs $\sim\log_2 N$ different turn-on and turn-off frequencies for the single nonresonant ac field in order to be able to approximate any unitary transformation with the accuracy of $1/N$th of the orbit. The result naturally applies to controlling wave packet revival structures.

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