

Two-color control of localization: From lattices to spin systems

Joanna Karczmarek,^{1,2} Malcolm Stott,² and Misha Ivanov¹

¹*Steacie Institute for Molecular Science, NRC, 100 Sussex Drive, Ottawa, Ontario, Canada K1A 0R6*

²*Physics Department, Queen's University, Kingston, Ontario, Canada K7L 3N6*

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We demonstrate control of quantum dynamics in a finite model system described by a tight-binding Hamiltonian, through interaction with a multifrequency external field. Effective defects can be introduced into the lattice by a two-frequency field, and the character of the defects can be controlled by the relative phase between the two field components. These field-induced defects imply robust localization of dressed (Floquet) states on lattice sites. Implications for a spin system in crossed magnetic fields are discussed. [S1050-2947(99)51211-0]

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The desire to control dynamics of a quantum system is both a driving force and a unifying theme in many areas of modern laser-matter interaction physics. For example, atom optics focuses on controlling the translational degrees of freedom of an atom. Molecular optics [1] strives to extend this ability to molecules. Coherent control [2] focuses on using laser fields to manipulate the internal dynamics of atomic and molecular systems; e.g., creating complex superpositions of quantum states according to a given prescription [3].

Following the experiment [4], coherent control in solid-state systems became an active area [5]. In quantum-well semiconductor structures, one of the objectives is the control of electron motion between quantum wells. An intriguing theoretical prediction is the coherent suppression of electron tunneling between the wells [6] by application of a strong monochromatic THz electric field. Following the first experiments in quantum-well structures [7], this idea has been transplanted back into the quantum-optics context: narrowing of Bloch bands has been observed for cold neutral atoms in optical lattices [8], with ac driving due to phase modulation of the counterpropagating waves that create the lattice.

Suppression of tunneling is caused by a destructive interference of different multiphoton quantum pathways, and has far reaching consequences in the case of the tight-binding lattice. For example, manipulation of the strength of the THz field allows control of the effective strength of the existing defects in the lattice, the Anderson localization length, and consequently affects the temperature dependence of electron transport [9].

We have studied numerically and analytically coherent control of quantum dynamics in a tight-binding model system subject to a multifrequency periodic external field, extending the monochromatic field study [9]. Although our analytical results apply to any periodic field, we focus on the simple case of only two frequencies ω and 2ω , where already we find a wealth of new effects.

First, unlike the single-frequency case, in a two-frequency field a delocalized initial state can be localized adiabatically at a single well, because the Floquet states of the driven system can be so localized. Second, the site where the electron is localized may be changed by adjusting the relative phase of the two frequencies. Third, compared to its single-frequency counterpart, the two-frequency localization is

stronger, and robust with respect to small changes in the external field strength. The two-frequency field introduces effective defects into the lattice, the strength of which depends on the relative phase of the two colors. For equal couplings between the sites the defects are induced at the ends of the lattice, but for couplings that are not all equal the defects are also induced within the lattice. In the presence of decoherence, the two-frequency driving can induce a tunneling current, whose direction is controlled by the relative phase between the frequencies.

The Hamiltonian for the tight-binding system together with an external time-dependent field is

$$\begin{aligned}\hat{H} &= \hat{H}_0 + V(t)\hat{N} \\ &= \sum_1^{N-1} \Omega_n(|n\rangle\langle n+1| + |n+1\rangle\langle n|) \\ &\quad + V(t)\sum_1^N n|n\rangle\langle n|,\end{aligned}\quad (1)$$

where N is the number of lattice sites, $|n\rangle$ is the state localized at the n th site and the Ω_n 's are couplings between adjacent sites. $V(t) = \mathcal{E}(t)d$ is periodic with period $T = 2\pi/\omega$. $V(t)$ arises from the interaction with an external electric field $\mathcal{E}(t)$, d being the well spacing.

The Hamiltonian (1) could describe an electron in a multiple quantum-well structure, or an ion in an optical lattice. For neutral atoms in an optical lattice analogous Hamiltonian is realized by phase-modulating the optical waves creating the lattice [8]. Equation (1) can also be used to represent the dynamics of circular Rydberg atomic states in a circularly polarized microwave field (the so-called Trojan states) [10], or the Zeeman effect in crossed magnetic fields. The coalescence of Zeeman lines in an oscillating magnetic field crossed with a constant magnetic field, observed almost 30 years ago [11], is formally equivalent [9] to the suppression of tunneling discovered in 1990s. We also note recent control experiments [12], where the time-dependent magnetic field was actively controlled to create a prescribed coherent superposition of Zeeman levels.

To explore the dynamics of the system described by Eq. (1), we first apply a unitary transformation $|\Psi(t)\rangle$

$=\hat{U}(t)|\Psi'(t)\rangle$, where $\hat{U}(t)\equiv\exp[-if_0^tV(t')dt'\hat{N}]$ and $\hat{N}\equiv\sum n|n\rangle\langle n|$. This is a direct analog of the Kramers-Henneberger transformation often used for studying atoms in strong high-frequency fields [13]: it gives the exact solution of the problem when $H_0=0$. The transformed Hamiltonian is

$$\hat{H}'=\sum_1^{N-1}\Omega_n(G(t)|n\rangle\langle n+1|+G^*(t)|n+1\rangle\langle n|), \quad (2)$$

where $G(t)\equiv\exp[-if^tV(t')dt']$. For a periodic $V(t)$, $G(t)=\sum_{k=-\infty}^{\infty}G_k\exp(-ik\omega t)$. If $V(t)$ includes a slowly varying envelope $f(t)$, such as we shall introduce later, to turn on the laser field, then G_k is replaced by $f(t)G_k$. The population of the n th well is $P_n\equiv|\langle n|\Psi(t)\rangle|^2=|\langle n|\Psi'(t)\rangle|^2$.

The Hamiltonian \hat{H}' describes N degenerate energy levels coupled by a multicolor ‘‘field’’ $G(t)$. Its time-independent part G_0 provides a resonant coupling, and dominates over the high-frequency, off-resonance couplings, $G_k\exp(-ik\omega t)$, which primarily cause energy shifts similar to ac Stark shifts. Following the usual procedure for ac Stark shifts, we eliminate the fast time dependence by treating terms such as $\Omega_n[G_k\exp(-ik\omega t)|n\rangle\langle n+1|+G_k^*\exp(ik\omega t)|n+1\rangle\langle n|]$ in second-order time-dependent perturbation theory. For example, the term $G_k^*\exp(ik\omega t)|n+1\rangle\langle n|$ gives an amplitude $\langle n+1|\Psi'(t)\rangle=-\langle n|\Psi'\rangle\Omega_nG_k^*\exp(ik\omega t)/k\omega$ in the state $|n+1\rangle$ which, when substituted back into $i\langle n|\Psi'(t)\rangle$, yields $-\langle n|\Psi'(t)\rangle\Omega_n^2G_kG_k^*/k\omega$. This diagonal contribution describes the energy shift.

When all the shifts are accounted for to second order we obtain the following time-independent effective Hamiltonian:

$$\begin{aligned} \hat{H}'_{eff} &= \sum_1^{N-1}\Omega_n[G_0|n\rangle\langle n+1|+G_0^*|n+1\rangle\langle n|] \\ &+ G_{\Sigma}\sum_1^N(\Omega_{n-1}^2-\Omega_n^2)|n\rangle\langle n|, \end{aligned} \quad (3)$$

where $G_{\Sigma}=\sum_{k\neq 0}G_kG_k^*/k\omega$, $\Omega_0\equiv\Omega_N\equiv 0$, and the result holds if $\omega\gg|G_k\Omega_n|$ for all n .

Of the same order as the ac Stark shifts are the ‘‘two-photon’’ Raman-type couplings between the degenerate states $|n\rangle$ and $|n+2\rangle$ due to $G(t)=\sum G_k\exp(-ik\omega t)$. These terms are absent from Eq. (3) because the corresponding matrix elements that enter to second order, $\Omega_{n,n+2}^{(2)}=\Omega_n\Omega_{n+1}\sum_{k\neq 0}G_kG_{-k}/k\omega$, vanish, as can be seen by changing the summation index from k to $-k$.

The stationary eigenstates $|\Psi'\rangle$ of H'_{eff} are approximations to the Floquet states of \hat{H}' and, hence, of \hat{H} (up to the phase-altering transformation \hat{U}). The dominant part of the rapid time dependence of $|\Psi\rangle=\hat{U}(t)|\Psi'\rangle$ (with the period of the driving field) is included in $\hat{U}(t)$. The relatively slow time dependence of $|\Psi'\rangle$, due, for instance, to the turning on or off of the laser field, is included in H'_{eff} by replacing G_k with $f(t)G_k$. We now discuss the rich physics contained in H'_{eff} and illustrate the effects with numerical examples.

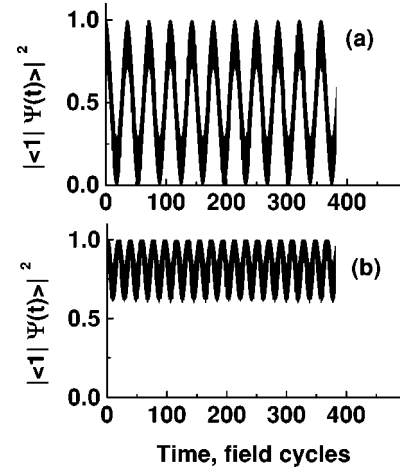


FIG. 1. One-frequency vs two-frequency localization for $N=2$ wells. Curves show population in the state $|n=1\rangle$. Initially $|\Psi(t=0)\rangle=|1\rangle$; the field turn-on is instantaneous. (a) Single frequency, $V_0/\omega=2.525$, $\Omega/\omega=1/4$. (b) Two frequencies, $V_0/\omega=2.60$, $\Omega/\omega=1/4$, $\phi=0$.

Equation (3) shows that the oscillating field $V(t)$ induces energy shifts Δ_n in the diagonal terms that are proportional to differences in the coupling strengths, $\Delta_n=G_{\Sigma}(\Omega_{n-1}^2-\Omega_n^2)$, breaking the degeneracy of the site energies. The distribution of shifts among the lattice sites is determined by Ω_n 's, but the overall magnitude and sign are controlled by the magnitude and sign of G_{Σ} , which are determined by $V(t)$. Furthermore, $V(t)$ modifies the couplings between the sites: $\Omega_n\rightarrow\Omega_nG_0$.

When $V(t)$ is such that G_0 is zero, H'_{eff} is diagonal, with eigenvalues, or quasienergies, $\tilde{E}_n=G_{\Sigma}(\Omega_{n-1}^2-\Omega_n^2)$ and eigenstates $|n\rangle$. The Floquet states of the dressed system become localized on lattice sites. Thus, coherent decoupling of lattice sites, or coherent destruction of tunneling between them, has been achieved.

For the case of a single-frequency field $V(t)=V_0\cos\omega t$, the condition $G_0=0$ for the coherent decoupling of lattice sites reduces to the well-known result [6] $J_0(V_0/\omega)=0$, where J_0 is the zero-order Bessel function. However, for a single-frequency field $G_{\Sigma}=0$, since $|G_k|=|G_{-k}|$. Consequently there are no diagonal energy shifts; the on-site energies remain degenerate. Tunneling is destroyed only at $G_0=J_0(V_0/\omega)=0$, and recovers as soon as G_0 deviates from zero, requiring exact tuning of V_0/ω .

In contrast, the addition of the second frequency, $V(t)=V_0[\cos(\omega t)+\cos(2\omega t+\phi)]$, ensures that $|G_k|\neq|G_{-k}|$ and so $G_{\Sigma}\neq 0$ (except at $\phi=\pm\pi/2$). This lifts the degeneracy of the site energies, creating energy defects in the lattice and making localization of the Floquet states robust. Localization persists as long as the energy shifts are large enough: $\Omega_n|G_0|<|G_{\Sigma}(\Omega_{n+1}^2-\Omega_n^2)|$.

This qualitative difference between the effects of single-frequency and two-frequency fields is illustrated in Fig. 1. The Schrödinger equation given by \hat{H} in Eq. (1) was integrated numerically to obtain the populations of the sites as functions of time. The results shown in Fig. 1 are for a double-well system with $\phi=0$. In a monochromatic field [Fig. 1(a)], a small 5% detuning of V_0 from the value at

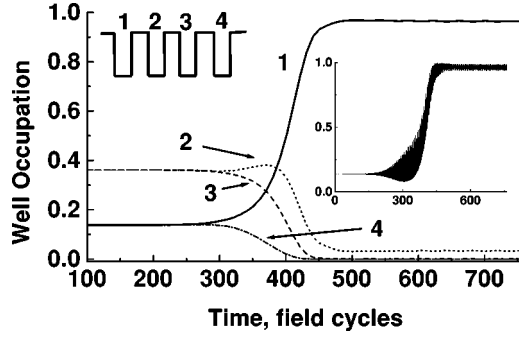


FIG. 2. Adiabatic localization in an $N=4$ -well system (sketched in the upper left corner). Curves show occupation in $|n\rangle$'s well, as numbered. Initially $|\Psi(t=0)\rangle$ is in the delocalized ground state of \hat{H}_0 . All Ω_n are equal, $\Omega_n=\Omega$; $V_0/\omega=2.48$, $\Omega/\omega=1/4$, $\phi=0$. For each point on the n th curve the occupations are averaged over ten cycles. Inset shows full time dependence for $|n=1\rangle$ before averaging, demonstrating the extent of fast oscillations in the occupation of the first well.

which $G_0=0$ [given by $J_0(V_0/\omega)=0$] destroys the localization completely: all population moves back and forth between the two wells. In the two-frequency field [Fig. 1(b)] the same 5% detuning of V_0 from the condition $G_0=0$ does not destroy the effect; localization is robust with respect to small changes in the field.

For nondegenerate Floquet states the system, starting in an eigenstate of H_0 , evolves adiabatically into a single Floquet state of H as the laser field $V(t)$ is slowly turned on. According to Eq. (3), the two-frequency field breaks the degeneracy of the Floquet states, and thus adiabatic localization of an electron around the induced defects is possible. Figure 2 shows numerical results for a four-well system with Ω_n the same for all n . V_0 is chosen so that $G_0=0$, making H'_{eff} diagonal. The initial state is the delocalized ground state of H_0 . As the field $V(t)=V_0[\cos(\omega t)+\cos(2\omega t+\phi)]$ is slowly turned on, Fig. 2 shows that the population is adiabatically localized in the leftmost (first) well for $\phi=0$. Localization occurs in the rightmost (fourth) well for $\phi=\pi$ (not shown).

Adiabatic localization can be achieved at sites for which the eigenstates of H'_{eff} are nondegenerate. Inspection of Eq. (3) shows that under the conditions used for Fig. 2, where all Ω_n are equal (and $G_0=0$), the eigenstates of H'_{eff} are degenerate, except for the leftmost well with an energy shifted down by $G_\Sigma\Omega$, and the rightmost well shifted up by $G_\Sigma\Omega$. These are the only wells where robust localization is possible for equal Ω_n . As the field is slowly turned on, the ground state of H_0 evolves into the lowest-energy Floquet state of H , which corresponds to the leftmost well when $G_\Sigma>0$. Changing the relative phase ϕ of the two frequencies from 0 to π reverses the sign of G_Σ and moves the lowest-energy Floquet state to the rightmost well.

If the set of Ω_n 's values is chosen suitably, localization can be engineered in any selected well. The spectrum of site energies in H'_{eff} is determined by (i) the n dependence Ω_n^2 , and (ii) the sign of G_Σ , which is reversed by changing ϕ from $\phi=0$ to $\phi=\pi$. For example, with $\Omega_n\propto(n-N/2)^2$ the second term in the Hamiltonian equation (3) depends linearly on n , as if there were a dc bias across the lattice. Changing the relative phase from $\phi=0$ to $\phi=\pi$ reverses the sign of

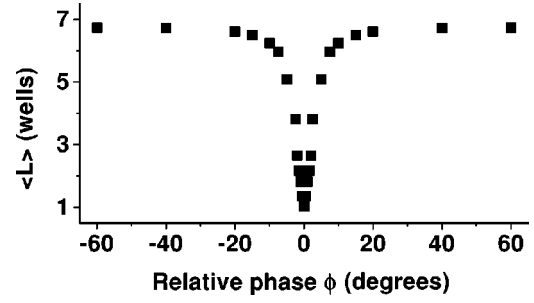


FIG. 3. Phase control of the Floquet states' localization length in an $(N=10)$ -well system with random couplings: $\Omega_n=0.5(1+\alpha)\Omega$; α is random between 0 and 1. $V_0/\omega=2.48$; $\Omega/\omega=1/4$.

G_Σ and, hence, the sign of the dc bias induced by the periodic field.

Our next example (Fig. 3) shows phase control of the localization length of the Floquet states and, hence, of quantum transport properties in a lattice with random couplings Ω_n . Figure 3 was calculated for $N=10$ wells with $\Omega_n=0.5(1+\alpha)\Omega$ and α random between 0 and 1. The localization length L of a state $|\psi_j\rangle$ is defined as $L_j=1/\sum_n|n|\psi_j|^4$, which gives $L=N$ for equal populations in each well. In Figure 3 we show the localization length averaged over all $N=10$ Floquet states (found numerically). For each of them $|n|\psi_j|^2$ is averaged over the field period. Bare states are strongly delocalized, with average localization length $\langle|L|\rangle=6.74$. In contrast, when the energy shifts induced by the applied field exceed the field-modified couplings between the wells, $\Omega_n|G_0|$, the Floquet states localize on single sites for $\phi=0, \pm\pi$. Changing ϕ controls $\langle|L|\rangle$ (Fig. 3).

One of many physical situations described by the tight-binding Hamiltonian in Eq. (1) is the Zeeman effect for a system with fixed total angular momentum J , such as an atom in a given electronic state, in crossed magnetic fields. We take the constant field B_x to be along the x axis, the oscillating field $B_z(t)$ to be along the z axis, and the states $|n\rangle$ to be the eigenstates of \hat{J}_z , with n ranging from $-J$ to J and $N=2J+1$. Consequently, $\hat{H}_0=g\mu_B B_x \hat{J}_x$, and $V(t)=g\mu_B B_z(t)$, where μ_B is the Bohr magneton and g is the Landé factor. The couplings, $\Omega_n=g\mu_B B_x \langle n|\hat{J}_x|n+1\rangle$, vary with n .

Following the method outlined above, the transformation $\hat{U}(t)=\exp[-i\hat{J}_z g\mu_B \int_0^t B_z(t') dt']$ is a time-dependent rotation. The new frame rotates back and forth about the z axis through an angle $\theta=g\mu_B \int B_z(t') dt'$. J_z is unaffected by this rotation. The transformed and effective Hamiltonians are

$$\hat{H}' = \frac{g\mu_B B_x}{2} [G(t)\hat{J}_- + G^*(t)\hat{J}_+], \quad (4)$$

$$\hat{H}'_{eff} = \frac{g\mu_B B_x}{2} (G_0\hat{J}_- + G_0^*\hat{J}_+) + G_\Sigma \frac{(g\mu_B B_x)^2}{2} \hat{J}_z,$$

$$G(t) = \exp\left(-ig\mu_B \int_0^t B_z(t') dt'\right),$$

where $\hat{J}_{\pm} \equiv \hat{J}_x \pm i\hat{J}_y$. In H'_{eff} the term proportional to \hat{J}_z arises from the difference in couplings, $\langle n|\hat{J}_x|n+1\rangle$. The term $G_0\hat{J}_- + G_0^*\hat{J}_+$ in H'_{eff} reduces to $2|G_0|\hat{J}_x$ after an additional rotation about the z axis by a constant angle θ_0 defined through $G_0 = |G_0|\exp(-i\theta_0)$. This rotation is made in the direction opposite to the previous one, and again, does not affect J_z . The resulting effective Hamiltonian takes a simple form $A\hat{J}_x + C\hat{J}_z$, with $A = g\mu_B B_x G_0$ and $C = G_{\Sigma}(g\mu_B B_x)^2/2$.

When $B_z(t) = 0, G_{\Sigma} = 0$; thus $C = 0$ and the eigenstates of the system have well-defined J_x . However, when the amplitude of $B_z(t)$ is such that $G_0 = 0$, A vanishes and J_z becomes a good quantum number, as reported [11] for a single-color case. In the two-color case J_z remains a good quantum number as long as the effective field along the x axis, $B_x^{(eff)} = |G_0|B_x$, is sufficiently small: $|G_0|g\mu_B B_x \ll G_{\Sigma}(g\mu_B B_x)^2$.

As with the lattice, we can adiabatically move the system from an initial eigenstate of J_x to an eigenstate of J_z by slowly turning on a suitably chosen $B_z(t)$. Since the expect-

ation value $\langle J_z \rangle$ is invariant under rotations about the z axis, and we control $\langle J_z \rangle$ in the rotating frame through the two-color field, we control $\langle J_z \rangle$ in the laboratory frame. For example, changing ϕ reverses the direction of $\langle J_z \rangle$.

So far we have neglected the effect of decoherence, which is important in quantum semiconductor structures. The key parameter should be the ratio of the field period T to the phase relaxation time T_{ph} . For $T \ll T_{ph}$ there is sufficient time to establish the Floquet states, and the relaxation will occur between these rather than the bare states [6,14]. If the magnitude of field-induced energy shifts, Δ_n exceeds the energy relaxation width T_{en}^{-1} , localization survives. In the opposite case of $\Delta_n T_{en} < 1$ an interesting situation arises if Ω_n are chosen to ensure that Δ_n depends linearly on n . Then, as we have seen, the two-color field introduces an effective dc bias that will induce a tunneling current. Its direction is reversed by changing the relative phase of the two frequencies ϕ from 0 to π . This complements phase control of ionization current first demonstrated in [4].

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- [1] See, e.g., H. Stapefeldt *et al.*, Phys. Rev. Lett. **79**, 2787 (1997); T. Takekoshi, B.M. Patterson, and R.J. Knize, *ibid.* **81**, 5105 (1998); A. N. Nikolov *et al.* (unpublished); T. Seideman, J. Chem. Phys. **106**, 2881 (1997); H. Sakai *et al.*, Phys. Rev. A **57**, 2794 (1998); B. Friedrich and D. Herschbach, Phys. Rev. Lett. **74**, 4623 (1995).
- [2] See, e.g., R. J. Gordon and S. A. Rice, Ann. Rev. Phys. Chem. **48**, 601 (1997); M. Shapiro and P. Brumer, J. Chem. Soc., Faraday Trans. **93**, 1263 (1997); H. Rabitz, Adv. Chem. Phys. **101**, 315 (1997).
- [3] T. Weinacht, J. Ahn, and P.H. Bucksbaum, Nature (London) **397**, 233 (1999).
- [4] E. Dupont *et al.*, Phys. Rev. Lett. **74**, 3596 (1995).
- [5] See, e.g., *Coherent Control in Atoms, Molecules, and Semiconductors*, edited by W. Pötz and A. Schroeder (Kluwer, Dordrecht, 1999).
- [6] See review by M. Grifoni and P. Haenggi, Phys. Rep. **304**, 229 (1998).
- [7] B.J. Keay *et al.*, Phys. Rev. Lett. **75**, 4102 (1995).
- [8] K.W. Madison *et al.*, Phys. Rev. Lett. **81**, 5093 (1998).
- [9] M. Holthaus, in *Coherent Control in Atoms, Molecules, and Semiconductors* (Ref. [5]), p. 171.
- [10] See, e.g., M. Kalinski and J. H. Eberly, Opt. Express **1**, 215 (1997), and references therein; I. Bialynicki-Birula, M. Kalinski, and J.H. Eberly, Phys. Rev. Lett. **73**, 1777 (1994); A.F. Brunello, T. Uzer, and D. Farrelly, *ibid.* **76**, 2874 (1996).
- [11] S. Haroche *et al.*, Phys. Rev. Lett. **24**, 861 (1970).
- [12] D. J. Heinzen and G. Xu (unpublished); see also G. Xu and D. Heinzen, Phys. Rev. A **59**, R922 (1999) for one-color experiments.
- [13] See, e.g., N. B. Delone and V. P. Krainov, *Multiphoton Processes in Atoms* (Springer-Verlag, Berlin, 1994).
- [14] O. Kocharovskaya, P. Mandel, and M.O. Scully, Phys. Rev. Lett. **74**, 2451 (1995).