PHYSICAL REVIEW A VOLUME 56, NUMBER 6 DECEMBER 1997

Controlling quantum coherence by circularly polarized fields

Jiushu Shao and Peter Hänggi
Institut für Physik, Universität Augsburg, Memminger Straße 6, D-86135 Augsburg, Germany

(Received 30 July 1997)

In terms of analytical results, we demonstrate that, within most parts in parameter space for a two-level magnetic system, suppression of quantum coherence can be induced by the *circularly polarized fields.* Low-frequency driving yields a dynamical localization opposite to the case of intermediate-to-high-frequency driving. The intrinsic relation between coherent suppression and the dressed-state level-crossing resonance is revealed. [S1050-2947(97)51112-7]

PACS number(s): 33.80.Be, 03.65.−w

By the principle of superposition, linear combinations of eigenstates are physically legitimate states. If the involved eigenstates are not all degenerate, then the superposed states vary with time. This effect is generally referred to as quantum coherence. As one can imagine, the simplest model exhibiting coherence is a two-level system (TLS). Such a TLS typifies many physical systems in diverse fields. For instance, it can model either exactly a spin-1/2 particle or approximately the chiral states of an optically active molecule. Moreover, a particle in a double-well potential can also be approximated as a TLS in the regime of low excitation energies. As a consequence, coherence and destruction of coherence in this case are better interpreted as tunneling and localization, respectively [1]. In the following, both tunneling and coherence will be used interchangeably.

Based on numerical calculations, coherent suppression of tunneling of a charged particle in a double-well potential was reported seven years ago [2]. The driving force was chosen to be a linearly polarized electric field with well-adjusted parameters that lie on a one-dimensional manifold [3]. Similar results were obtained for the linearly polarized drive TLS that approximately describes the double-well system [3–5]. Many applications resulting from this discovery are, among others [6], the laser-induced trapping of an electron in a quantum-well structure and the control of electron transfer reactions [7–11]. According to Hund's theory, the change of chirality, or an intramolecular rearrangement in a more general sense, may be regarded as a tunneling process of a (charged) particle in a double-well potential [12]. Therefore, one can employ appropriate electric fields to localize the particle, hence stabilizing one configuration of the molecule.

How about localization for a circularly polarized field (CPF) driving? Let us consider a spin-1/2 particle of magnetic dipole moment μ in a constant field B_z. The Hamiltonian is $H_0 = -\Delta_0 \sigma^z/2$, where $\Delta_0 = \mu B_z$. The eigenstates of the system are denoted by $|1\rangle$ and $|2\rangle$. With a driving circularly polarized (magnetic) field whose direction rotates in the plane perpendicular to z axis, the Hamiltonian thus reads

$$H(t) = -\frac{1}{2}\Delta_0 \sigma^z + \frac{V_0}{2}[\sigma^+ \exp(i\omega t) + \sigma^- \exp(-i\omega t)], \tag{1}$$

where $V_0 = -\mu B$ and $\sigma^\pm = \sigma^x \pm i\sigma^y$. Localization in a TLS has been investigated for the case of linearly polarized driving fields [3–5]. Can an appropriate CPF also induce destruction of coherence in a TLS? If yes, what is the mechanism underlying localization? We will answer these questions from both dynamical and quasienergy points of view. The effect of quantized fields will also be addressed.

First let us explain the mathematical structure of localization for a periodically driven TLS. The generic Hamiltonian is

$$\hat{H} = \begin{pmatrix} -\Delta_0/2 & V(t) \\ V^*(t) & \Delta_0/2 \end{pmatrix}, \tag{2}$$

where $V(t)$ is a periodic function of time, i.e., $V(t+T) = V(t)$. In the following we set $\hbar = 1$. The wave function $|\psi(t)\rangle$ can be written as

$$|\psi(t)\rangle = c_L(t)|L\rangle + c_R(t)|R\rangle, \tag{3}$$

where $|L\rangle = (|1\rangle + |2\rangle)/\sqrt{2}$ (left state) and $|R\rangle = (|1\rangle - |2\rangle)/\sqrt{2}$ (right state) are the two superposed (localized) states of interest. The system is exactly characterized by the vector

$$\mathbf{C}(t) \equiv \begin{pmatrix} c_L(t) \\ c_R(t) \end{pmatrix}.$$

From the Schrödinger equation $i\partial|\psi(t)\rangle/\partial t = H(t)|\psi(t)\rangle$, one readily derives

$$\frac{d\mathbf{C}(t)}{dt} = M(t)\mathbf{C}(t), \tag{4}$$

where

$$M(t) = i\left(\frac{1}{2}\Delta_0 \sigma^x + \mathrm{Im}\{V(t)\}\sigma^y - \mathrm{Re}\{V(t)\}\sigma^z\right).$$

This is a linear dynamical system with periodic coefficients. We can employ Arnold's geometric approach to study the time-evolution dynamics [13]. To this end we define $\mathbf{A}(t) \equiv \mathcal{T}\exp[\int_0^t dt' M(t')]$, where $\mathcal{T}$ is the time-ordering operator. The most useful property is

$$\mathbf{C}(nT+t) = \mathbf{A}(t)\mathbf{A}^n \mathbf{C}(0),$$

where $\mathbf{A}\equiv\mathbf{A}(T)$ is the T-advancing map. Because $M(t)$ is anti-Hermitian, $\mathbf{A}(t)$ is unitary, such that

$$\mathbf{A}=\begin{pmatrix} a & -c \\ c^* & a^* \end{pmatrix}, \tag{5}$$

where $|a|^2+|c|^2=1$.

Although there is not a general definition, by localization we mean that the probability of finding the system in the initial state will always stay large. Noting that $\mathbf{A}$ is a unimodular matrix, we can use the Caley-Hamilton theorem (see, e.g., [14]) to obtain

$$\mathbf{A}^n=P_{n-1}(\alpha)\mathbf{A}-P_{n-2}(\alpha)\mathbf{1}, \tag{6}$$

where P is the Chebyshev polynomial, $P_n(\alpha)=\sin[(n+1)\alpha]/\sin\alpha$, with $\alpha=\arccos(\mathrm{Tr}\mathbf{A}/2)$ and $\mathbf{1}$ is the identity matrix. Let the system initially evolve from the left state $|\psi(0)\rangle=|L\rangle$. Localization requires that $c_R(t)$ be always small. Consider stroboscopic times; using Eq. (6), one obtains

$$|c_R(nT)|=\frac{|c|}{\sqrt{[\mathrm{Im}(a)]^2+|c|^2}}|\sin(n\alpha)|. \tag{7}$$

Localization results from $|c|\ll|\mathrm{Im}(a)|$. This is a trivial consequence because this condition means that one of the two localized states has much less energy (localization via bias). Nontrivial localization occurs whenever $\sin(n\alpha)$ is small for any n, being possible if and only if $\sin(\alpha)=0$. Therefore we have $\alpha=\pm m\pi$ (m integer) [5]. In this case, both $|c_L(t)|^2$ and $|c_R(t)|^2$ evolve periodically [5].

Let us build the time-advancing map $\mathbf{A}(t)$ for the Hamiltonian given by Eq. (1). Because it can be solved explicitly [15], we will derive $\mathbf{A}(t)$ from the propagator $\mathcal{K}(t,t_0)$: $|\psi(t)\rangle=\mathcal{K}(t,t_0)|\psi(t_0)\rangle$, instead of by solving the dynamical system itself. Using a gauge transformation $|\widetilde{\psi}(t)\rangle=\mathcal{U}|\psi(t)\rangle$, where $\mathcal{U}=\exp(-i\omega t\sigma^z/2)$, we arrive at [16]

$$i\frac{\partial|\widetilde{\psi}(t)\rangle}{\partial t}=i\frac{\partial\mathcal{U}}{\partial t}|\psi(t)\rangle+i\mathcal{U}\frac{\partial|\psi(t)\rangle}{\partial t}=\widetilde{H}|\widetilde{\psi}(t)\rangle, \tag{8}$$

where

$$\widetilde{H}=\mathcal{U}H\mathcal{U}^\dagger+i\frac{\partial\mathcal{U}}{\partial t}\mathcal{U}^\dagger=-\left(\frac{\Delta_0}{2}-\frac{\omega}{2}\right)\sigma^z+V_0\sigma^x$$

is *time independent*. The energy eigenvalues are $\widetilde{\epsilon}_\pm=\pm\sqrt{(\Delta_0-\omega)^2/4+V_0^2}$. Noting that $|\psi(t)\rangle=\mathcal{U}^\dagger|\widetilde{\psi}(t)\rangle=\mathcal{U}^\dagger\exp(-i\widetilde{H}t)|\widetilde{\psi}(0)\rangle$, we have

$$\mathcal{K}(t,0)=\mathcal{U}^\dagger\exp(-i\widetilde{H}t).$$

The quantum map $\mathbf{A}(t)$ is nothing but a representation of the propagator in the basis of $|L\rangle$ and $|R\rangle$. After some algebra we find

$$\mathbf{A}(t)=\begin{pmatrix} a(t)-ib(t) & -c(t)+id(t) \\ c(t)+id(t) & a(t)+ib(t) \end{pmatrix}, \tag{9}$$

where

$$a(t)=\cos(\Omega t/2)\cos(\omega t/2)+\cos\theta\sin(\Omega t/2)\sin(\omega t/2),$$

$$b(t)=\sin\theta\sin(\Omega t/2)\cos(\omega t/2),$$

$$c(t)=\sin\theta\sin(\Omega t/2)\sin(\omega t/2),$$

$$d(t)=\cos(\Omega t/2)\sin(\omega t/2)-\cos\theta\sin(\Omega t/2)\cos(\omega t/2)$$

with $\Omega=\sqrt{(\Delta_0-\omega)^2+4V_0^2}$ being the Rabi frequency and $\tan\theta=2V_0/(\omega-\Delta_0)$.

We readily calculate the T-advancing map $\mathbf{A}$ over a single period $T=2\pi/\omega$. Recall the general treatment given above. The necessary condition for localization is $\arccos(\mathrm{Tr}\{\mathbf{A}(T)\}/2)=m\pi$. Because $\mathrm{Tr}\{\mathbf{A}(2\pi/\omega)\}=-2\cos(\Omega\pi/\omega)$, we obtain $\Omega/\omega=n$ (n any positive integer). The probability of the system in the left state $|L\rangle$ is

$$\begin{aligned}P_L(t)&=a^2(t)+b^2(t)\\&=\frac{1}{2}[1+\sin^2\theta\cos(\omega t)+\cos\theta\sin(\omega t)\sin(n\omega t)\\&\quad+\cos^2\theta\cos(\omega t)\cos(n\omega t)].\end{aligned} \tag{10}$$

We now prove that localization is possible only for $n=1$. To this end let us consider the time average of P_L, denoted as $\overline{P_L}$. Simple algebra yields

$$\begin{aligned}\overline{P_L}&=\frac{\omega}{2\pi}\int_0^{2\pi/\omega}dt[a^2(t)+b^2(t)]\\&=\frac{1}{2}+\frac{\sin(2n\pi)}{4\pi(n^2-1)}(n\cos^2\theta+\cos\theta).\end{aligned} \tag{11}$$

One readily finds $\overline{P_L}=1/2$ for all $n\neq1$. That is, the system is always delocalized if the Rabi frequency is a higher-order multiple of the driving frequency $\Omega=n\omega$, $n>1$. Therefore, localization may take place only if the Rabi frequency coincides with the driving frequency. For $n=1$, then, we thus find the very relationship between the period and the amplitude of the acting field:

$$\Omega=\omega=\frac{\Delta_0^2+4V_0^2}{2\Delta_0}. \tag{12}$$

This is the central result of the CPF driving. With this condition we find for $P_L(t)$ and $\overline{P_L}$, respectively,

$$\begin{aligned}P_L(t)=\frac{1}{2}\Bigg\{1&+\frac{x^2-1}{x^2+1}\sin^2(\omega t)+\cos(\omega t)\\&\times\left[\frac{4x^2}{(x^2+1)^2}+\frac{(x^2-1)^2}{(x^2+1)^2}\cos(\omega t)\right]\Bigg\}\end{aligned} \tag{13}$$

and

$$\overline{P_L}=\frac{1+x^2+2x^4}{2(1+x^2)^2}, \tag{14}$$

where $x=2V_0/\Delta_0$. A plot of $\overline{P_L}$ vs x is depicted in Fig. 1.

On the one hand, absolute localization, $\overline{P_L}\to1$, will be realized for $x\to\infty$. This requires an infinitely strong field

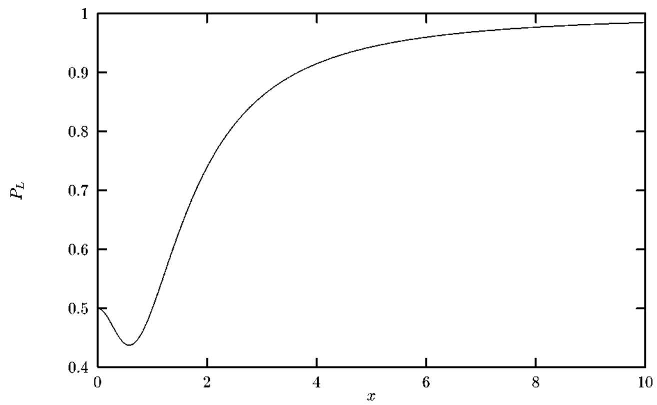

FIG. 1. Time average of the probability $\overline{P_L}$ of the system remaining in the initial state $|L\rangle$ as a function of $x=2V_0/\Delta_0$ under the localization condition.

with an infinite high frequency, as determined by Eq. (12). On the other hand, strong localization will be achieved already for intermediate-to-strong fields, because $\overline{P_L}$ rapidly assumes the saturation value one (see Fig. 1). For instance, setting $V_0=2\Delta_0$ and $\omega=17\Delta_0/2$ (i.e., $x=4$), we find $\overline{P_L}>0.91$. For $x=4$, a continuous time evolution of P_L is shown in Fig. 2.

An interesting effect appears for $x<1$. In this case, the system slightly tends to occupy the right state $|R\rangle$, although it initially evolves from the left state $|L\rangle$. The minimum of $\overline{P_L}$ is

$$\overline{P_L^{min}}=\frac{7}{16}, \tag{15}$$

which corresponds to

$$\omega^{min}=\frac{2}{3}\,\Delta_0, \quad V_0^{min}=\frac{1}{2\sqrt{3}}\,\Delta_0 \quad \left(x=\frac{1}{\sqrt{3}}\right).$$

Figure 2 also displays the variation of $P_L^{min}(t)$ with time.

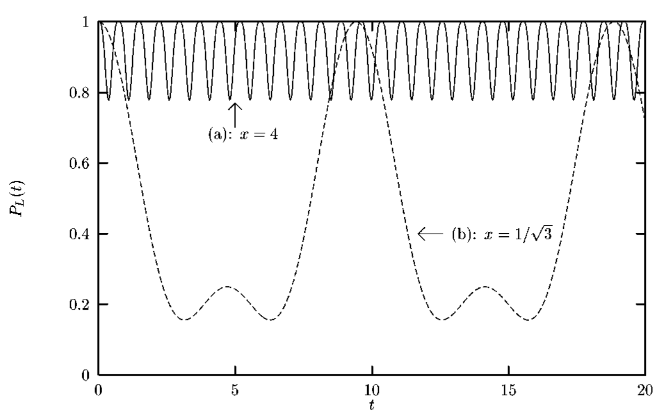

FIG. 2. Time evolution of the probability of staying in the left localized state $P_L(t)$ $[P_L(0)=1]$ under the localization condition, Eq. (12): (a) strong localization corresponding to $x\equiv 2V_0/\Delta_0=4$ and $\omega/\Delta_0=17/2$; (b) weak localization in the other state, $P_L^{min}(t)$, corresponding to $x=1/\sqrt{3}$ and $\omega/\Delta_0=2/3$. Dimensionless time is measured in terms of Δ_0; $t\rightarrow t(\Delta_0/\hbar)\equiv t$.

From the consideration of time-dependent dynamics, it has been demonstrated that the CPF may induce localization. We next use the Floquet theory to reveal the localization mechanism [3]. The theory is based on the eigenvalue problem of the Hermitian operator $\mathcal{H}(t)\equiv\hat{H}-i\partial/\partial t$:$\mathcal{H}(t)\Phi_\alpha(t)=\epsilon_\alpha\Phi_\alpha(t)$. The eigenfunctions are called Floquet modes and the corresponding eigenvalues are called quasienergies. One can show that the Floquet mode is periodic, i.e., $\Phi_\alpha(t)=\Phi_\alpha(t+T)$ and the quasienergy ϵ_α is unique up to multiples of the driving frequency ω [16]. The quasienergies in our model are

$$\epsilon_{\pm,n}=[\tilde{\epsilon}_\pm+(n+1/2)\omega] \mod\omega.$$

The condition of localization $\Omega=\omega$ coincides with the exact level crossing: $\epsilon_{+,0}=\epsilon_{-,1}$. This may be understood as follows. When driven by the periodic CPF, the two eigenstates are surrounded by the field to become the dressed states with the quasienergies as their energies. As in the bare case, when the level crossing happens, coherence is suppressed. Real localization is expected when the level crossing takes place frequently in the manifold of the dressed system. It should be stressed again that *the level crossing of the quasienergies yields a necessary (but not sufficient) criterion for suppression of coherence* [3]. Therefore, just as for the linearly polarized case [3], a similar interference mechanism applies for the circularly polarized field to induce coherent suppression of tunneling.

Concerning the effect of a quantized field, the corresponding Hamiltonian reads

$$H=-\frac{1}{2}\,\Delta_0\sigma^z+\omega a^\dagger a+g(a\sigma^-+a^\dagger\sigma^+), \tag{16}$$

where g is the coupling constant, which is assumed to be real for simplicity. This is the one-mode Jaynes-Cummings model in quantum optics and it exhibits several interesting features such as collapse and revival [17]. Recognizing that the interaction couples only the TLS-field states $|1,n+1\rangle\equiv|1\rangle\otimes|n+1\rangle$ and $|2,n\rangle\equiv|2\rangle\otimes|n\rangle$ for each value of n, one can readily evaluate the energy eigenvalues

$$E_{\pm n}=(n+1/2)\omega\pm\Omega_n/2.$$

The corresponding eigenvectors or dressed states are

$$|+n\rangle=\sin\,\theta_n|1,n+1\rangle+\cos\,\theta_n|2,n\rangle,$$

$$|-n\rangle=\cos\,\theta_n|1,n+1\rangle-\sin\,\theta_n|2,n\rangle,$$

where θ_n is determined by

$$\tan\,2\theta_n=\frac{2g\sqrt{n+1}}{\Delta_0-\omega}.$$

If $E_{+n-1}=E_{-n}$, then level-crossing resonances take place [18]. In the semiclassical limit $n\rightarrow\infty$, one identifies $g\sqrt{n}=V_0$ and $\Omega_n=\Omega$, so that from $E_{+n-1}=E_{-n}$ one again finds $\Omega=\omega$. Therefore, the observable level-crossing-resonance reflects suppression of quantum coherence. The same conclusion can be drawn for the linearly polarized fields [18–20].

The CPF can choose one of two helicities, which correspond to positive and negative ω. One can show that only the CPF with positive ω is able to induce localization. This is not surprising because only the clockwise CPF (i.e., the rotating-wave terms of the linearly polarized field drive) interacts effectively with the TLS. We should also point out that localization within our characterization is a quasistatic property (long-time average). It does not necessarily mean a reduction of tunneling rate.

In summary we have demonstrated that—as in the case of linearly polarized fields—appropriate circularly polarized fields can induce suppression of quantum coherence in a driving two-level magnetic system; cf. Eqs. (12)–(14). The condition is that the Rabi frequency coincide with that of the applied circularly polarized field. Strong suppression can be achieved for intermediate-to-strong fields (compared to the energy splitting of the two-level system). If the driving field is weak, however, the system slightly tends to occupy the other localized state. It should be pointed out that preparation of a superposed (localized) state is in general a subtle problem [7]. For a spin-1/2 magnetic particle placed in a constant field B_2, however, this can be realized by using a strong static magnetic field (in a direction different from the z axis) that is suddenly switched off before the circularly polarized field is turned on. The localization effect is closely related to the dressed-state level-crossing resonance and may be tested by experiments [18].

J.S. would like to thank W. H. E. Schwarz for useful discussions. This work was supported by the Alexander von Humboldt-Stiftung, the National Natural Science Foundation of China, and the Deutsche Forschungsgemeinschaft (P.H.) via the Schwerpunktprogramm ''*Zeitabhängige Phänomene und Methoden in Quantensystemen der Physik und Chemie*'' (Grant No. Az. HA 1517/14-2).

[1] Rigorously speaking, tunneling is associated with continuum states while coherence is associated with discrete states; cf. L. A. Leggett, in *Directions in Condensed Matter Physics,* edited by G. Grinstein and G. Mazenko (World Scientific, Singapore, 1986), Vol. I, pp. 187–248.

[2] F. Großmann, P. Jung, T. Dittrich, and P. Hänggi, Z. Phys. B **84**, 315 (1991); F. Großmann, T. Dittrich, P. Jung, and P. Hänggi, Phys. Rev. Lett. **67**, 516 (1991); F. Großmann, T. Dittrich, and P. Hänggi, Physica B **175**, 293 (1991).

[3] F. Großmann and P. Hänggi, Europhys. Lett. **18**, 571 (1992).

[4] J. M. Gomez Llorente and J. Plata, Phys. Rev. A **45**, R6958 (1992).

[5] L. Wang and J. Shao, Phys. Rev. A **49**, R637 (1994).

[6] P. Hänggi, in *Quantum Dynamics of Submicron Structures,* edited by H. A. Cerdeira, B. Kramer, and G. Schön (Kluwer Academic, Dordrecht, 1995), p. 673.

[7] R. Bavli and H. Metiu, Phys. Rev. Lett. **69**, 1986 (1992); Phys. Rev. A **47**, 3299 (1993).

[8] M. Morillo and R. I. Cukier, J. Chem. Phys. **98**, 4548 (1993).

[9] D. E. Makarov and N. Makri, Phys. Rev. E **52**, 5863 (1995).

[10] Y. Dakhnovskii, V. Lubchenko, and R. D. Coalson, Phys. Rev. Lett. **77**, 2917 (1996).

[11] M. Grifoni, L. Hartmann, and P. Hänggi, Chem. Phys. **217**, 167 (1997).

[12] F. Hund, Z. Phys. **43**, 805 (1927).

[13] V. I. Arnold, *Ordinary Differential Equations* (MIT Press, Cambridge, MA, 1973); *Mathematical Methods of Classical Mechanics* (Springer-Verlag, New York, 1978); *Geometric Theory of Ordinary Differential Equations* (Springer, Berlin, 1983).

[14] M. O. Vassell, J. Lee, and H. F. Lockwood, J. Appl. Phys. **54**, 5206 (1983).

[15] I. I. Rabi, Phys. Rev. **51**, 625 (1937).

[16] P. Hänggi, in T. Dittrich, P. Hänggi, G.-L. Ingold, B. Kramer, G. Schön, and W. Zwerger, *Quantum Transport and Dissipation* (VCH, Weinheim, 1997), Chap. 5.

[17] See, for instance, P. Meystre and M. Sargent III, *Elements of Quantum Optics,* 2nd ed. (Springer, Berlin, 1991), Chap. 13.

[18] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Atom-Photon Interactions* (Wiley, New York, 1992), Chap. 6.

[19] J. Plata and J. M. Gomez Llorente, Phys. Rev. A **48**, 782 (1993).

[20] P. Neu and R. J. Silbey, Phys. Rev. A **54**, 5323 (1996).