Conditions leading to intense low-frequency generation and strong localization in two-level systems

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(Received 30 November 1992)

A charged particle in a double well can emit, when driven by an intense laser, intense low-frequency radiation or become strongly localized in one of the wells. By using perturbation theory and an asymptotic analysis, we determine the conditions for which these processes take place for a two-level model.

PACS number(s): 42.50.−p

The behavior of a charged particle in a symmetric double well driven by a strong laser has been studied recently in several articles [1–7]. Grossman, Dittrich, Jung, and Hanggi [1,2] have shown that the laser can prevent a particle located initially in one of the wells from tunneling to the other well. Bavli and Metiu [8,9] demonstrated that a semi-infinite Gaussian pulse can take the electron from a delocalized energy eigenstate of the double well, localize it in one of the wells, and keep it there.

The emission properties of an electron in a double well are also interesting. Numerical solutions of the time-dependent Schrödinger equation [7] show that the Fourier transform $\mu(\Omega)$ of the induced dipole $\mu(t)$ has peaks at three types of frequencies: shifted even harmonics (SEH) ($\Omega=2n\omega\pm\Delta$ with $n=1,2,\ldots$), pure odd harmonics [$\Omega=(2n+1)\omega$, with $n=0,1,2,\ldots$], and the frequency $\Omega=\Delta$. The shift Δ is a function of the laser frequency ω and its intensity I. These parameters can be chosen to make Δ arbitrarily small and $\mu(\Omega)$ has a very-low-frequency peak. The presence of this peak was called [7] low-frequency generation (LFG). For certain values of $\{\omega,I\}$, called [7] points of accidental degeneracy (AD), Δ becomes equal to zero and $\mu(t)$ acquires a static component. In a symmetric double well the presence of a *static* dipole, induced and maintained by the laser, is possible only if the electron density has a static part that is asymmetrically distributed between the wells, i.e., if electron localization occurs.

The calculations also show [7] that when $\{\omega,I\}$ approaches an AD point the LFG intensity is sometimes very large, exceeding the intensity of the fundamental (i.e., the component having the frequency $\Omega=\omega$); as $\{\omega,I\}$ reaches an AD point the system has, sometimes, a very large static dipole. This means that the laser-induced localization is strong.

Finally, it is also observed that if $\{\omega,I\}$ is not at an AD point then $\mu(\Omega)$ has no even harmonics (EH) (i.e., no peaks at $\Omega=2n\omega$, $n=1,2,\ldots$), in agreement with the selection rules which state that such peaks are forbidden to all orders in perturbation theory. As $\{\omega,I\}$ approaches an AD point and $\Delta\to 0$, the SEH peaks (at the frequencies $\Omega=2n\omega\pm\Delta$) get closer and closer and become pure even harmonics; strong even harmonic generation is observed, in disagreement with the selection rules.

Clearly the shift Δ is a central parameter in describing this phenomenology; interesting processes take place when Δ is *small* or *zero*. In this article we use a combination of perturbation theory and asymptotic analysis to provide a simple formula for Δ and to find the conditions under which the LFG is intense and the localization is strong.

To do this we assume that the processes described above can be represented in the space spanned by the two lowest-energy eigenstates $|1\rangle$ and $|2\rangle$ of the bare (i.e., no radiation) Hamiltonian. Bavli [10] validated this assumption by showing that the time-dependent wave function for the double-well model used in Refs. [6] and [7] can be represented at all times, with better than 90% accuracy, by the linear combination $a_1(t)|1\rangle+a_2(t)|2\rangle$.

We use the two-level Hamiltonian

$$H=\varepsilon\{|1\rangle\langle 1|-|2\rangle\langle 2|\}-\{|1\rangle\langle 2|+|2\rangle\langle 1|\}\mu_{12}E(t)$$
$$=\varepsilon\sigma_z-\mu_{12}E(t)\sigma_x \ . \tag{1}$$

Here $|2\rangle$ and $|1\rangle$ are the ground and first excited states of the electron in the double well, respectively, and σ_x and σ_z are Pauli matrices. The zero of energy is halfway between the energy levels ε_1 and ε_2, and $2\varepsilon=\varepsilon_1-\varepsilon_2$.

The induced dipole is

$$\mu(t)=\mu_{12}\langle\Psi,t|\sigma_x|\Psi,t\rangle \ , \tag{2}$$

where $|\Psi,t\rangle$ satisfies the time-dependent Schrödinger equation with the Hamiltonian (1). By using standard methods we derive for $\mu(t)$ the equation of motion

$$\frac{d\mu(\tau)}{d\tau}=-(\varepsilon/\hbar\omega)^2\left\{\cos[e_0\sin(\tau)]\int_0^\tau d\tau'\mu(\tau')\cos[e_0\sin(\tau')]+\sin[e_0\sin(\tau)]\int_0^\tau d\tau'\mu(\tau')\sin[e_0\sin(\tau')]\right\} , \tag{3}$$

with $e_0=2\mu_{12}E_0/\hbar\omega$ and $\tau=\omega t$. By using [11]

$$\exp[ie_0\sin(\tau)]=\sum_{n=-\infty}^{\infty}J_n(e_0)\exp[in\tau] \ , \tag{4}$$

where J_m are Bessel functions of integer order, we can rewrite Eq. (3) as

$$\frac{d\mu(\tau)}{d\tau}=-[\varepsilon J_0(e_0)/\hbar\omega]^2\int_0^\tau d\tau'\mu(\tau')-(\varepsilon/\hbar\omega)^2F(\tau;\mu)\ , \tag{5}$$

with

$$F(\tau;\mu)=2\sum_{n=1}^{\infty}J_{2n}(e_0)\cos[2n\tau]\int_0^\tau d\tau'\mu(\tau')\left[J_0(e_0)+2\sum_{k=1}^{\infty}J_{2n}(e_0)\cos[2n\tau']\right]$$
$$+2\sum_{n=1}^{\infty}J_{2n+1}(e_0)\sin[(2n+1)\tau]\int_0^\tau d\tau'\mu(\tau')\left[2\sum_{k=0}^{\infty}J_{2k+1}(e_0)\cos[2n\tau']\right]\ , \tag{6}$$

Equation (6) can be formally "solved" to give

$$\mu(\tau)=\mu_{12}\cos[\Delta\tau/\omega]-(\varepsilon/\hbar\omega)^2\int_0^\tau d\tau'\cos[\Delta(\tau-\tau')]F(\tau';\mu)\ . \tag{7}$$

We identify the frequency

$$\Delta=[\varepsilon J_0(e_0)/\hbar] \tag{8}$$

appearing in the first term in the right-hand side with the shift Δ discussed earlier in this article. Therefore, the AD points are the points in the (ω,I) plane given by $2\mu_{12}E_0/\hbar\omega=r_n$, where r_n are the zeros of J_0. In going from (5) to (7) we have used the initial condition $\mu(t=0)=\mu_{12}$ corresponding to an initially localized state. The behavior of the system with the initial condition $\mu(t=0)=0$ is very different [12] from the one found here; there is no low-frequency generation and at the AD points the system is completely delocalized.

In the zeroth-order approximation (i.e., if $\varepsilon/\hbar\omega\rightarrow 0$) Eq. (7) gives $\mu(t)=\mu_{12}\cos[\Delta t]$ and the Fourier transform $\mu(\Omega)$ has a peak at the frequency Δ. When Δ is small this corresponds to low-frequency generation. In this approximation the intensity of the LFG peak is very high, as compared to other peaks, since μ_{12} is the highest amplitude a Fourier component can have.

We must now examine the first-order correction $\mu_1(t)$ and try to answer several questions. Will the higher-order terms modify the frequency Δ? Will they change the amplitude μ_{12} of the zeroth-order term? Under what conditions is the zeroth-order term larger than all the others?

The first-order correction is

$$\mu_1(\tau)=-(\varepsilon/\hbar\omega)^2\int_0^\tau d\tau'\cos[\tau-\tau']F(\tau';\mu_0)\ . \tag{9}$$

This is obtained by replacing $\mu(\tau)$ under the integral in Eq. (7) with μ_0. The analysis of this term is tedious and we present here only the conclusions. (i) We find no indication that the first- or the higher-order terms will renormalize the shift Δ. Floquet theory [9] shows that in a two-level system there could be only one shift frequency [7]. Therefore, for all parameters for which the perturbation expansion is valid the shift is given by Eq. (8). (ii) The first-order correction contains terms whose time dependence is given by $\cos[\Delta t]$. As a result, the corrected dipole has the form $\mu(t)=[\mu_{12}-(\varepsilon/\hbar\omega)^2a]\cos\Delta t$, where a is a sum of Bessel functions squared and is always smaller than one. Thus as long as

$$\varepsilon/\hbar\omega\ll 1\ , \tag{10}$$

this amplitude correction is small. (iii) $\mu^{(1)}$ contains shifted and pure harmonic terms. The amplitude of these terms is of the form $(\varepsilon/\hbar\omega)^2b$, where b is less than one; as long as Eq. (10) is satisfied the amplitude of these terms is smaller than μ_0. (iv) The first-order corrections contain "secular" terms of the form $t(\varepsilon/\hbar\omega)^2\Delta\sin(\Delta t)$, which become infinitely large in time. This is a frequent nuisance in the time-dependent perturbation theory. It does not mean that $\mu(t)$ diverges, but only that as we increase t we are stepping outside the radius of convergence of the expansion. Thus we are certain that $\mu_{12}\cos(\Delta t)$ is the dominant contribution in $\mu(t)$ only if Eq. (10) and

$$t(\varepsilon/\hbar\omega)^2\Delta\ll 1 \tag{11}$$

are satisfied. Note that Eq. (11) implies that the time when the secular terms become important is longer and longer as $\Delta\rightarrow 0$; there are no secular terms if $\Delta=0$.

The condition (11) implies that it is possible that LFG is intense and the localization is strong only for a finite time, even if $(\varepsilon/\hbar\omega)^2<1$. Our numerical experiments found no evidence for this, which suggests that when calculated to all orders, the secular terms add up to a well-behaved function whose values are smaller than μ_0.

To see under what conditions the secular terms will lead to corrections that are smaller than μ_0 we have performed an asymptotic analysis of $\mu(t)$. We start from

$$\mu(\tau)/\mu_{12}=1-(\varepsilon/\hbar\omega)^2\sum_{n=-\infty}^{+\infty}\sum_{k=-\infty}^{+\infty}J_n(e_0)J_k(e_0)\int_0^\tau d\tau_1\exp[in\tau_1]\int_0^{\tau_1}d\tau_2\exp[-in\tau_2]\mu(\tau_2)/\mu_{12}\ ,$$

which is obtained by integrating Eq. (3), then using Eq. (4). This equation can be solved by successive iterations. The first-order term in the iteration scheme is obtained by putting $\mu(\tau_2)=1$ under the integral in the right-hand side. Performing the integrals leads to an expression of the form

$$1-(\varepsilon/\hbar\omega)^2[J_0(e_0)^2(\tau)^2/2+\tau d+g+f(\tau)] \ .$$

Here d and g are smaller than one and $f(t)$ contains sines and cosines of $n\omega t$, where n is an integer. As the iteration proceeds the term $1-(\varepsilon/\hbar\omega)^2 J_0(e_0)^2\tau^2/2$ leads to $\cos[(\Delta/\omega)t]$. This cosine will exceed the next fastest growing secular term if $\tau J_0(e_0)/2 >> d$. For the series leading up to the cosine to converge, we must also have

$$\tau(\varepsilon/\hbar\omega)J_0(e_0)<1 \ . \tag{12}$$

These two conditions give the time interval in which $\cos(\Delta t)$ is the largest secular term in the expansion

$$\tfrac{1}{2} << \omega t J_0 < 1/(\varepsilon/\hbar\omega) \ . \tag{13}$$

If J_0 is very small the asymptotic analysis ensures that $\mu_{12}\cos(\Delta t)$ is the dominant term in $\mu(t)$ at long times; moreover, condition (8) ensures this dominance for short times. Together, these conditions cover the whole territory. They state that if $(\varepsilon/\hbar\omega) \ll 1$ and if Δ is small we have intense LFG; if $(\varepsilon/\hbar\omega) \ll 1$ and $\Delta=0$ we have good localization.

Bavli and Metiu [8] tested these results by solving the equation of motion for $\mu(t)$ numerically. In Fig. 1 we show $\mu(t)$ for $(\varepsilon/\hbar\omega) \ll 1$, when the predictions made by the theory are expected to work. We find that if the parameters are such that Eq. (8) predicts that $\Delta=0$ the calculated $\mu(t)/\mu_{12}$ is equal to a constant which is close to one, plus a term oscillating with a small amplitude and a high frequency. $\mu(t)/\mu_{12}$ is equal to one at all times only

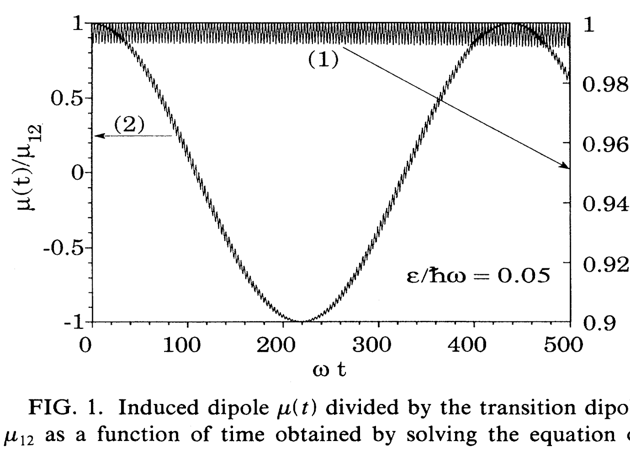

FIG. 1. Induced dipole $\mu(t)$ divided by the transition dipole μ_{12} as a function of time obtained by solving the equation of motion numerically. (1) $e_0=2\mu_{12}E_0/\hbar\omega=2.4$, which corresponds to a point of accidental degeneracy [i.e., Δ given by Eq. (8) is zero], hence to localization; (ii) $e_0=2\mu_{12}E_0/\hbar\omega=2.7$. The population oscillates in accordance with Eq. (7). Since $\varepsilon/\hbar\omega \ll 1$, $\mu(t)$ is dominated by the first term in Eq. (7).

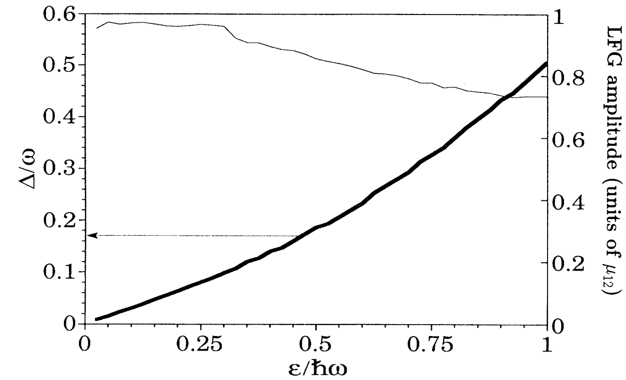

FIG. 2. Dependence of the shift frequency and of the amplitude $\mu(\Delta)$ of the low-frequency mode on $\varepsilon/\hbar\omega$ for $2\mu_{12}E_0/\hbar\omega=6.0$.

if the electron is perfectly localized, at all times, in one of the wells. The example shown in Fig. 1 comes very close to this. We found no case in which the predictions of the theory were in error. Figure 1 also shows a case in which $(\varepsilon/\hbar\omega) \ll 1$ and the value of Δ given by Eq. (8) is small. The time evolution of $\mu(t)$ shows a large-amplitude–low-frequency component and a high-frequency–low-amplitude one; as predicted by theory the term $\mu_{12}\cos(\Delta t)$ dominates the behavior of $\mu(t)$ and LFG is intense. We have also calculated the Fourier transform of $\mu(t)$ and obtained from it the value of Δ and the absolute value of the Fourier component $\mu(\Delta)$. We varied the laser frequency and the laser field intensity E_0 so that $e_0=2\mu_{12}E_0/\hbar\omega=6$. The theory predicts that as long as $(\varepsilon/\hbar\omega) \ll 1$ Eq. (8) is valid and Δ is proportional to $(\varepsilon/\hbar\omega)$. The plot shows this to be true. The deviations from this prediction are not large even for $(\varepsilon/\hbar\omega)$ of order 2. Figure 2 also shows the absolute value of $\mu(\Delta)/\mu_{12}$ which is the intensity of LFG. As long as $(\varepsilon/\hbar\omega) \ll 1$ this is close to one which is the maximum intensity for this model. In a double quantum well μ_{12} is rather large, leading thus to large LFG emission. The predictions of the present theory also work well for almost all the AD points discovered numerically in Ref. [7]. The few discrepancies occur probably because the system studied in Ref. [7] is approximately a two-level system.

Previous work [9,10] has used perturbation theory to calculate the energy difference ΔE between the Floquet quasienergies. LFG and SHG were not discussed, but the connection to localization was made [1,2]: localization can occur only if the Floquet quasienergies are equal; in the present context this means $\Delta=0$. However, previous work used the high-field limit and obtained ΔE under more restrictive conditions than those given here. The question of how large a fraction of the electron density is localized and how intense LFG is, to our knowledge, has not been addressed before. We find that these effects are large if $(\varepsilon/\hbar\omega) \ll 1$. While the field strength is important for making Δ small, it is not a player in determining whether the LFG is intense or the localization is strong.

While we used quantum wells as an example, the two-level analysis presented here is more general and these processes may be detectable for other systems. For example, Corkum and co-workers [13] have observed them while exposing ions to a strong short pulse.

This work was supported by NSF Grant No. CHE91-12926. We are very grateful to Raanan Bavli for performing the numerical calculations reported here and to Martin Holthaus for useful discussions.

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