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Symmetry-breaking effects induced by intense laser fields

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We consider the interaction of an intense laser field with an ensemble of electrons confined by a symmetric potential well, and show that, for times shorter than the dephasing time, the laser field can break the symmetry of the system regardless of its initial conditions, resulting in optical rectification and even-harmonic generation. In particular, we show that relatively moderate fields can give rise to significant even-harmonic generation, even if the system is initially in thermal equilibrium, with no need for pulse shaping, suggesting that the detection of symmetry-breaking effects may be experimentally accessible.

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The problem of localization of an electron in a symmetric double quantum well by an intense laser pulse has been studied extensively in recent years [1–3]. In the presence of a strong driving field, an electron, initially localized in one well, will tunnel back and forth between the wells at a frequency equal to the transition frequency between the Floquet states of the system [4]. For certain values of frequency and intensity of the driving field for which the Floquet states become degenerate, the tunneling process is suppressed and the initial localization is maintained. If, in addition to frequency and intensity, the rise time of a smoothly rising pulse acting on the system is chosen appropriately, it is possible not only to maintain initial localization but even to localize an electron initially in a delocalized state [2,3]. In this work we show that even-harmonic generation occurs for arbitrary initial conditions and conclude that initial localization and pulse shaping are not required for symmetry breaking, although they can greatly enhance it in the extremely-strong-field regime. The emission spectrum of the dressed system exhibits several interesting features. There are two groups of spectral lines: odd harmonics of the incident field, and shifted even harmonics at frequencies $2k\omega\pm\tilde{\omega}$, where k is zero or integer, ω is the frequency of the driving field, and $\tilde{\omega}$ is the transition frequency between the two Floquet states of the system. Pure even-harmonic generation occurs when the Floquet states become degenerate ($\tilde{\omega}=0$). Thus an intense laser pulse with appropriate parameters breaks the symmetry of the system.

In this Rapid Communication we generalize the single-electron treatment adopted in previous work to an incoherent ensemble of electrons confined by a symmetric potential well. We assume that at time $t=0$ the laser is turned on abruptly, and study the response of the system to the field for different initial conditions, by solving the density-matrix equations. We restrict our analysis to a two-level system [2,3]. We distinguish between two cases. In the first case the system is taken to be initially in a state of coherent superposition of localized electron wave packets (localized state), and in the second one the system is initially in thermal equilibrium. We find that in both cases a large driving field with specific frequency and intensity breaks the symmetry of the system. However, while in the former case the direction favored by the system is determined by the initial state of the system, in the latter case it is determined by the direction of the electric field at the turn-on time. We also find that for extremely strong laser fields, even-harmonic generation is strongly enhanced if the system is initially in a localized state, but is insensitive to the initial state of the system for moderate fields. We show that localization and symmetry breaking are transient phenomena and persist only for times shorter than the dephasing time. Finally, we discuss the possibility of observing symmetry-breaking effects experimentally.

The density-matrix equations describing the dynamics of the system under consideration can be written in the form

$$\frac{dw}{dt}=-4\Omega v\cos(\omega t)-\frac{w-w_{\mathrm{eq}}}{T_1}, \quad (1)$$

$$\frac{dv}{dt}=\omega_{12}u+\Omega w\cos(\omega t)-\frac{v}{T_2}, \quad (2)$$

$$\frac{du}{dt}=-\omega_{12}v-\frac{u}{T_2}. \quad (3)$$

Here $w=\rho_{22}-\rho_{11}$ is the population inversion, w_{eq} is the value of w at equilibrium (taken to be -1 in what follows), $v=(\rho_{12}-\rho_{21})/2i$, $u=(\rho_{12}+\rho_{21})/2$ is the dipole moment of the system, $\Omega=Ed_{12}/\hbar$ is the Rabi frequency, E is the amplitude of the incident field, ω_{12} is the transition frequency between the states of the bare system, ω is the frequency of the laser, and T_1 and T_2 are the relaxation and dephasing times, respectively. At very high intensities, $\Omega\gg\omega_{12}$, the above system can be solved by using perturbation theory with $\omega_{12}/\sqrt{\omega\Omega}$ serving as the smallness parameter [3]. It is convenient to transform to the new variables:

$$\alpha=w\cos\eta+v\sin\eta, \quad \beta=-w\sin\eta+v\cos\eta, \quad (4)$$

where $\eta=(2\Omega/\omega)\sin(\omega t)$. The density-matrix equations then take the form

$$\frac{d\alpha}{dt}=\omega_{12}u\sin\eta-\frac{\alpha+\cos\eta}{T}, \tag{5}$$

$$\frac{d\beta}{dt}=\omega_{12}u\cos\eta-\frac{\beta-\sin\eta}{T}, \tag{6}$$

$$\frac{du}{dt}=-\omega_{12}(\beta\cos\eta+\alpha\sin\eta)-\frac{u}{T}, \tag{7}$$

where, for simplicity, we have assumed that $T_1=T_2=T$. The first term on the right-hand side of Eqs. (6) and (7) contains the constant term $J_0(2\Omega/\omega)$ which gives rise to linear growth of the first-order corrections, where J_0 is the zeroth-order Bessel function. To avoid this problem, this term must be taken into account exactly [3].

We first consider the case in which the system is initially in a state of coherent superposition of localized electron states; that is, we assume that the charge of the system is initially localized on one of the sides of the system $[u(t=0)\neq0]$. To zeroth order the dipole moment $u^{(0)}$ has a low-frequency component,

$$u^{(0)}=e^{-t/T}[\beta(0)\sin(\tilde{\omega}t)+u(0)\cos(\tilde{\omega}t)], \tag{8}$$

where $\beta(0),u(0)$ are the initial conditions and $\tilde{\omega}=\omega_{12}J_0(2\Omega/\omega)$ is the transition frequency between the two Floquet states [1–3]. Evidently, the strong driving field gives rise to coherent oscillations of the electrons at a frequency $\tilde{\omega}$. When the parameter $2\Omega/\omega$ is equal to one of the zeros of the Bessel function J_0, $\tilde{\omega}$ vanishes and $u^{(0)}=e^{-t/T}u(0)$; that is, the dipole has a large static component directed along its initial $(t=0)$ direction. This means that the symmetry of the system is entirely broken by the strong field. It also means that, for a time shorter than T, the system "remembers" its initial conditions. The fact that the dipole moment is dominated by a large static component implies that the charge is kept localized by the laser.

As seen from Eq. (8) the low-frequency oscillations decay exponentially on a time scale T. The reason is that dephasing processes destroy phase coherence, thereby giving rise to destructive interference of the different Floquet states of individual electrons. Thus, after a time T the system loses its memory and the symmetry is restored.

The calculation of the first-order correction is tedious but straightforward. For simplicity we restrict our attention to the special case $T\to\infty$. The first-order term $u^{(1)}$ can then be written as a sum over odd-order harmonics of the incident field,

$$u_{2k+1}=-2\alpha(0)\frac{\omega_{12}}{\omega}\frac{J_{2k+1}(2\Omega/\omega)}{2k+1}\times\{\cos[(2k+1)\omega t]-\cos(\tilde{\omega}t)\}, \tag{9}$$

and shifted even harmonics,

$$u_{2k}=-\frac{\omega_{12}}{\omega}[u(0)\sin(\tilde{\omega}t)+\beta(0)\cos(\tilde{\omega}t)]\times\frac{J_{2k}(2\Omega/\omega)}{2k}\sin(2k\omega t). \tag{10}$$

The resonances at frequencies $2k\omega\pm\tilde{\omega}$ result from multiphoton processes associated with transitions between Floquet states. Note that the odd-order term also involves low-frequency oscillations. However, if the initial dipole moment $u(0)$ is of order $\alpha(0)$, the amplitude of the latter is smaller, by a factor $(\omega_{12}/\omega)(\omega/2\Omega)^{1/2}$, than the amplitude of the zeroth-order term, and therefore negligible. The general solution, which includes damping, is lengthy and will not be presented here. Basically, we find that when damping is included the even-harmonic terms decay exponentially much like the zeroth-order term. The odd-harmonic oscillations do not decay.

The initial conditions assumed above are not very realistic. Most likely, the system will be found initially in its equilibrium state, in which case $u(0)=\beta(0)=0$. Under these conditions the zeroth-order term $u^{(0)}$ and the even-harmonic terms u_{2k} given by Eqs. (8) and (10) vanish. Using Eq. (9), the dominant contribution to the low-frequency Fourier component of the dipole moment can then be expressed as $d\cos(\tilde{\omega}t)$, where

$$d=2\alpha(0)\frac{\omega_{12}}{\omega}\sum_{k=1}^{\infty}\frac{J_{2k+1}(2\Omega/\omega)}{2k+1}, \tag{11}$$

so that for $\tilde{\omega}=0$ we again obtain a static dipole. The important consequence is that even if the system is initially in an equilibrium state the symmetry of the system will be broken by a sufficiently strong field. However, the amplitude of the low-frequency oscillations appears to be of the same order as that of the odd-harmonic oscillations. This suggests that no localization will occur if the system is initially in equilibrium. The question now is: what is the direction favored by the system in this case? Close examination of the constant d, given by the last equation, reveals that the direction of the static dipole is determined by the direction of the electric field at the turn-on time, $t=0$, via the arguments of the Bessel functions. It can be easily shown that the phase of d is changed by π when the direction of the electric field at $t=0$ is reversed $(\Omega\to-\Omega)$.

To second order we obtain

$$u^{(2)}=-d\frac{\omega_{12}}{\omega}\sum_{k=1}^{\infty}\frac{J_{2k}}{2k}[\cos(2k\omega+\tilde{\omega})t-\cos(2k\omega-\tilde{\omega})t]. \tag{12}$$

As seen from the last equation the phase of the modes $2k\omega+\tilde{\omega}$ and $2k\omega-\tilde{\omega}$ differs by π. Therefore, when $\tilde{\omega}=0$ the two modes interfere destructively, implying that pure even-harmonic generation is completely inhibited to this order. The third-order correction,

$$u^{(3)}=\frac{d}{2}\left[\frac{\omega_{12}}{\omega}\right]^2\sum_{k,l=1}^{\infty}J_{2k}J_{2l}\left\{\frac{\sin\{[2(k+l)\omega+\tilde{\omega}]t\}}{4k(l+1)+\tilde{\omega}}+\frac{\sin\{[2(k-l)\omega+\tilde{\omega}]t\}}{4k(k-l)+\tilde{\omega}}+(\tilde{\omega}\to-\tilde{\omega})\right\}+(\text{odd-harmonic terms}), \tag{13}$$

comprises the pure even harmonics (when $\tilde{\omega}=0$). The inclusion of damping leads to an exponential decay of the even-harmonic terms to all orders.

We see that, for very strong fields, even-harmonic generation is strongly depressed if the system is initially in an equilibrium state. More precisely, the intensities of the shifted even harmonics and the pure even harmonics are reduced by factors $\omega_{12}/\sqrt{\omega\Omega}$ and $(\omega_{12}/\sqrt{\omega\Omega})^2$, respectively, relative to the intensity of the odd harmonics. One anticipates that, for moderate fields ($\Omega \simeq \omega \simeq \omega_{12}$), the intensity of the second harmonic peak may be comparable to the intensity of the Rayleigh peak (the intensity of the higher harmonic peaks will be strongly diminished). Furthermore, from Eqs. (12) and (13) we infer that each double line ($2k\omega \pm \tilde{\omega}$) will have an asymmetric structure. However, the validity of the perturbative solution is questionable for moderate fields. To study the response of the system to moderate fields, and also as a check on the analytic solution, we have solved Eqs. (1)–(3) numerically. Once the dipole moment $u(t)$ is obtained, we calculate its Fourier transform $u(\omega')$. We find that the analytic solution is in a fairly good agreement with the numerical one. Typical results of the numerical integration are shown in Figs. 1 and 2. In these examples the system was taken to be initially in equilibrium [$u(0)=v(0)=0$; $w(0)=-1$].

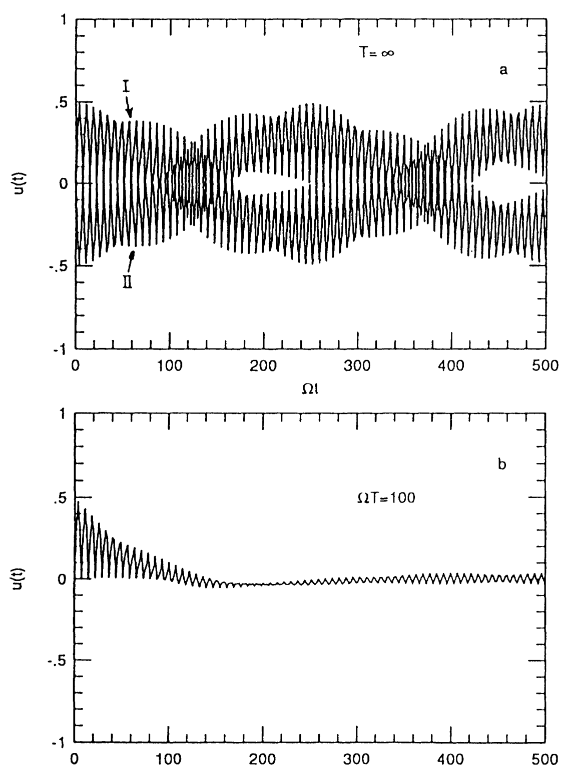

FIG. 1. (a) Time evolution of the dipole moment induced by a moderate laser field (see definition in the text) in a two-level system initially in equilibrium for two opposite directions of the electric field (I and II) at $t=0$ and for an infinite dephasing time. The time is given in units of the Rabi time. $\omega_{12}=0.5\Omega$ and $\omega=0.85\Omega$. (b) The effect of finite dephasing time on the evolution of the dipole moment. The parameters are the same as in (a).

Figure 1(a) shows the polarization obtained for two opposite directions of the electric field at the turn-on time. Obviously, the preferred direction is determined by the initial direction of the electric field. We emphasize that, though the phase of the dipole moment depends on the initial direction of the electric field, the existence of the even-harmonic peaks is independent of the initial phase of the field. The effect of damping is illustrated in Fig. 1(b). We find that in all cases the low-frequency and even-harmonic oscillations decay on a time scale T_2. Figure 2 presents the Fourier transform of the dipole moment. In both cases the spectrum has peaks at $\omega'=(2k+1)\omega$. The asymmetry of the double line structures $2k\omega\pm\tilde{\omega}$ is clearly seen Fig. 2(a). In Fig. 2(b) the parameter $2\Omega/\omega$ is approximately equal to the first zero of J_0. The height of the peak at $\omega'=2\omega$ is roughly $\frac{1}{3}$ the height of the Rayleigh peak. To check how different initial conditions affect the spectrum, we integrated the equations using the same parameters as in Fig. 2, but for different initial conditions. We find that the spectrum is insensitive to initial conditions. For example, for $u(0)=0.5$ the height of the peaks is changed only by a factor of order unity. The situation changed dramatically for much stronger fields. In that case the spectrum appears to be very sensitive to the initial conditions, in agreement with the analytical solution.

Our results, derived for a two-level system, will be modified by the presence of other energy levels. When the two interacting levels are well separated from the rest

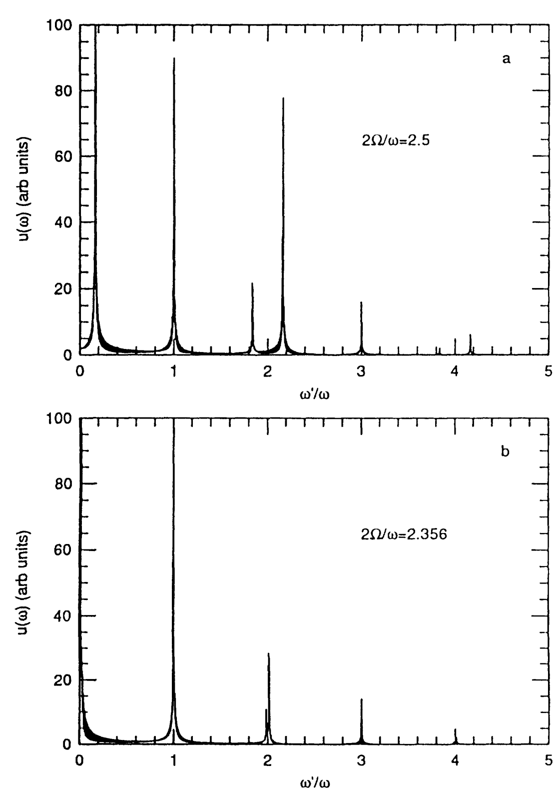

FIG. 2. Fourier transform of the dipole moment induced by a moderate field. The system is initially in equilibrium. $\omega_{12}=0.5\Omega$; $T=\infty$.

(e.g., double quantum well) the two-level approximation is fairly accurate. When the interaction with the other levels becomes significant, the spectrum may exhibit additional features, but the essential symmetry breaking will prevail.

We now discuss the feasibility for experimental observation of the above predicted phenomena. We consider intersubband transitions in the GaAs/Ga-Al-As quantum well, for two wavelength regimes: $\lambda = 10$ μm [5] and $\lambda \simeq 50$ μm [6]. In the first case, the regime of moderate fields can be reached for laser intensities in excess of 5×10^8 W/cm^2. The typical dephasing time, which limits the temporal window for observation of even harmonics, is between 100 fsec (at room temperature [7]) and 260 fsec (at temperatures below 100 K [8]), corresponding to 3 and 60 optical cycles, respectively. In the second case, the limitations are far less restrictive. The regime of moderate fields can be reached for intensities of the order 5×10^6 W/cm^2, and the corresponding dephasing time may be larger, due to the absence of longitudinal optical phonon interactions [9]. Thus phenomena associated with symmetry breaking may be detectable experimentally. It is worth noting that according to recent measurements [10], the maximum light power that a GaAs structure can withstand is above these current requirements for pulsed laser operation. However, the transient nature of even-harmonic generation implies that the experiments should be performed with short pulses of light of duration less than T_2. This may introduce a new direction of using a pulse train for both: even-harmonic generation and probing of the system in a manner similar to photon-echo experiments [11]. The regime of multipulse operation is currently under investigation.

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