Optimal Stochastic Enhancement of Photoionization

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(Received 28 November 2006; published 20 April 2007)

The effect of noise on the nonlinear photoionization of an atom due to a femtosecond pulse is investigated in the framework of the stochastic Schrödinger equation. A modest amount of white noise results in an enhancement of the net ionization yield by several orders of magnitude, giving rise to a form of quantum stochastic resonance. We demonstrate that this effect is preserved if the white noise is replaced by broadband chaotic light.

DOI: 10.1103/PhysRevLett.98.160201 PACS numbers: 02.50.Ey, 32.80.Rm, 42.50.Hz

The interplay between noise and the nonlinear response of a physical system described by classical mechanics has led to intriguing effects. A paradigm is stochastic resonance (SR), whereby a nonlinear system responds more strongly to coherent subthreshold driving in the presence of an appropriate amount of noise [1–3]. Despite prominent demonstrations of classical SR in ample variety, only a few quantum mechanical examples have been studied, possibly due to subtle features of the quantum evolution [3]. The concept of SR in the *quantum* domain was originally suggested in the context of two-level conductance switching of driven mesoscopic metals [4]. This proposition sparked a number of theoretical studies on quantum stochastic resonance in double-quantum well structures [5], the so-called incoherent and coherent SR in driven dissipative spin-boson systems [6], in the bistable photon-field dynamics in micromaser [7], and in the electron shelving in a single ion [8]. Experiments using NMR spectroscopy of water [9], and a very recent one in the radio-frequency controlled nanomechanical beam oscillator [10], have established the properties of quantum SR in two-level systems (TLS). These studies are mostly restricted to the quantum analog of the classical double-well dynamics. Yet, they have provided valuable insight into the noise-induced dynamics of quantum systems.

Following quite a different path of research, there is a rapidly growing activity in the general area of controlling quantum phenomena [11]. A common approach to exercise the control exploits the *nonperturbative* interplay between a purposefully designed optical field from a laser and the target quantum system, such as an atom or molecule [12]. A fundamental control goal is the manipulation of atomic or molecular interaction to steer the quantum system towards a desired state [11,12]. An accurate knowledge of the effect of noise on quantum systems would be very helpful to achieve full control. Their response to noise has been rarely studied so far [13]. One may even wonder whether the presence of noise offers new possibilities of quantum control.

Here, we will demonstrate the existence of a stochastic resonance-like effect in a generic quantum situation beyond the two-level systems. For this purpose, we consider a quantum system having a finite binding potential with mixed, discrete, and continuous spectrum which is coupled to two external forces: First, a nonresonant coherent optical driving and, second an incoherent perturbation which may result from some form of environment. These situations are sufficiently general to be achieved in a variety of quantum systems such as in nuclear motion of diatomic molecules, in Josephson junction devices, and in active single-electron atoms.

Let us concentrate on the latter example in the form of the simplest single-electron atom, i.e., the hydrogen atom. Because of the application of a linearly polarized laser field $F(t)$, the electron dynamics is effectively confined to one dimension along the polarization axis. The Hamiltonian for such a simplified (yet reliable [14]) description of the hydrogen atom, which is here also perturbed by a stochastic force $\xi(t)$, reads as (atomic units, $\hbar = m = e = 1$, are used unless stated otherwise)

$$H(x, t) = \frac{\hat{p}^2}{2} + V(x) + x\{F(t) + \xi(t)\}, \tag{1}$$

where x is the position of the electron and $\hat{p} = -i\partial/\partial x$ is the momentum operator. The binding potential is approximated by a nonsingular Coulomb-like form $V(x) = -1/\sqrt{x^2 + a^2}$. Such a soft-core binding potential with parameter a has been routinely employed to study atomic dynamics in strong laser fields [15]. It successfully describes many experimental features of multiphoton or tunnel ionization [14], and the observation of the plateau in the higher harmonic generation spectra [15]. The external perturbations (last term in Eq. (1)) is dipole coupled to the atom. The laser field is a nonresonant femtosecond pulse (duration 20 optical periods) described as $F(t) = f(t)F_0 \sin(\omega t + \delta)$. Here, $f(t)$ defines a smooth pulse envelope with F_0 and ω denoting peak amplitude and angular frequency, respectively. The noise term $\xi(t)$ is a zero-mean $\langle\xi(t)\rangle = 0$, Gaussian white noise with autocorrelation function

$$\langle\xi(t)\xi(t')\rangle = 2D\delta(t - t'), \tag{2}$$

and intensity D [16].

Because of the stochastic nature of the Hamiltonian, the quantum evolution is nondeterministic. Thus, an averaging

over a large number of realizations of the stochastic force is required in order to produce a statistically meaningful solution of the time-dependent stochastic Schrödinger equation

$$i\frac{\partial\Psi(x,t)}{\partial t} = H(x,t)\Psi(x,t). \quad (3)$$

For a given realization, the numerical solution of the Schrödinger equation amounts to propagating the initial wave function $|\Psi_0\rangle$ using the infinitesimal short-time stochastic propagator, $U_\xi(\Delta t) = \exp[-i\int_t^{t+\Delta t} H(x,t)dt]$. One can compute $U_\xi(\Delta t)$ using the standard FFT split-operator algorithm [17], with the stochastic integration in the exponential interpreted in the Stratonovitch sense [16]. Successive applications of the propagator $U_\xi(\Delta t)$ advance $|\Psi_0\rangle$ forward in time. Note that the initial state $|\Psi_0\rangle$ is the *ground state* of the system having a binding energy of $I_b = -0.5$ a.u.. This is obtained by the imaginary-time relaxation method for $a^2 = 2$ [14]. To avoid parasitic reflections of the wave function from the grid boundary, we employ an absorbing boundary [17]. The observable, such as the ionization flux leaking in the continuum on one side, is defined as $J_R(x_R, t) = \mathrm{Re}[\Psi^* \hat{p}\Psi]_{x_R}$, where x_R is a distant point (typically 500 a.u.) near the absorbing boundary. The ionization rate is integrated over a sufficiently long time interval to obtain the corresponding total ionization probability, $P = \int_0^\infty J_R(x_R, t)dt$.

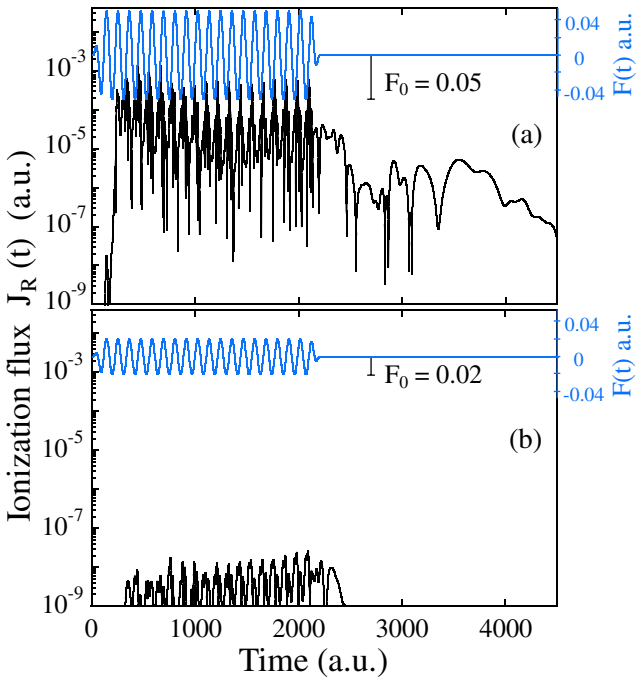

FIG. 1 (color online). Nonlinear ionization flux $J_R(t)$ (lower part of plots) induced by a 20 optical cycles laser pulse shown in the respective upper plots with $\omega = 0.057$, $\delta = 0$ and peak amplitude, $F_0 = 0.05$ (a), and $F_0 = 0.02$ (b). The pulse envelope $f(t)$ is unity except single-cycle sinusoidal rising and falling edges. The pulse has a narrow spectral width $\Delta\omega_{\mathrm{pulse}} = 0.0028$. The threshold for over barrier ionization is $F_{\mathrm{th}} = 0.067$ a.u..

First, we will discuss the response of the atom interacting with a short but strong laser pulse only. It produces (nonlinear) ionization of the atom which is most easily understood, especially in the time domain, with the picture of tunneling ionization. The periodic bursts of the ionization flux are produced close to those times when the effective potential $U(x,t) = V(x) + xF(t)$ is maximally bent down by the dipole-coupled laser field. Such temporal evolution of the ionization flux for a 20 cycle laser pulse is illustrated in Fig. 1 (shown in the top parts) with two different peak amplitudes $F_0 = 0.05$ a.u. [Fig. 1(a)] and $F_0 = 0.02$ a.u. [Fig. 1(b)].

Time-resolved ionization peaks separated by the optical period $(2\pi/\omega)$ are clearly visible for both peak field amplitudes. In addition, $J_R(t)$ shows a complex interference pattern due to the modulated Coulomb barrier in Fig. 1(a). However, quite strikingly, if F_0 is reduced to 0.02 a.u., the

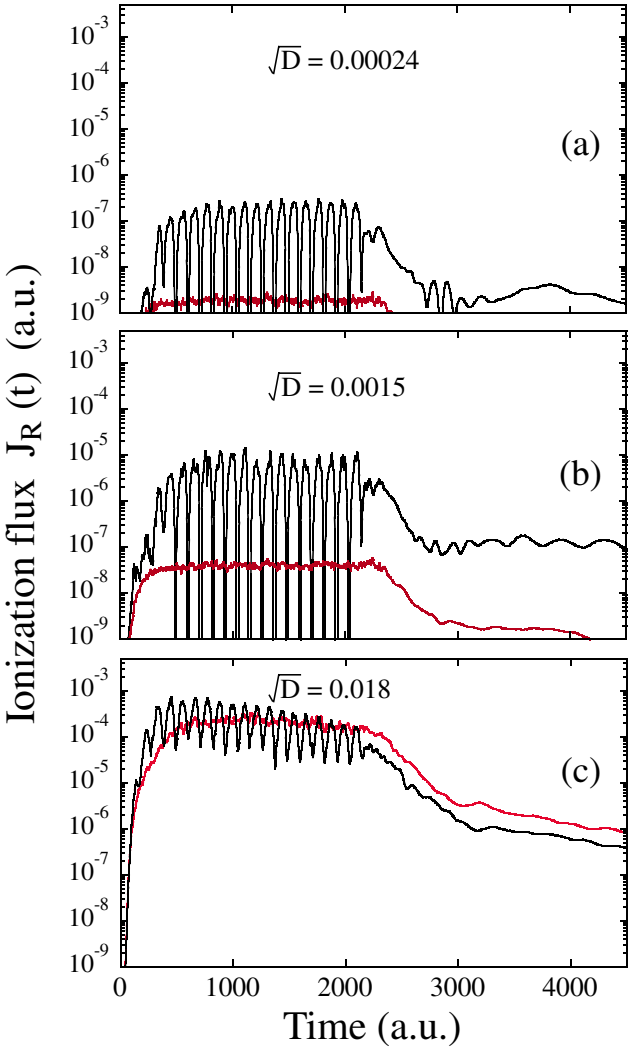

FIG. 2 (color online). Ionization flux for a weak laser pulse $F_0 = 0.02$, with three values of noise amplitude $\sqrt{D}$ (a) 0.00024, (b) 0.0015, and (c) 0.018. Background featureless curves show the corresponding purely noise-driven ($F_0 = 0$) flux. The flux is averaged over 50 realizations.

ionization collapses by around 5 orders of magnitude as shown in Fig. 1(b). One can conclude that the photoionization dynamics is highly nonlinear, and, in particular, it exhibits a form of "threshold" dynamics where the threshold is created by the condition for over barrier ionization.

Almost nothing is known about the quantum analog of SR in such "threshold-crossing" quantum devices [2]. Naturally, in the context of SR, the question arises if noise can recover the strong periodic ionization flux for the "weak" laser pulse. To answer this question, we show the ionization flux averaged over 50 realizations in Fig. 2(a) when a small amount of noise is added to the weak laser pulse ($F_0 = 0.02$). Note that the noise is switched on for the same time interval as the 20 cycle laser pulse. One can see that for very small noise amplitude $\sqrt{D} = 0.00024$, the periodic structure in atomic ionization gets amplified by 1 order of magnitude as compared to the case of the laser pulse alone [compare Fig. 1(b) with Fig. 2(a)]. However, one might ask if such an enhancement could be due to the noise alone. Contrary to the coherent excitation, the noise alone produces an, on average, featureless ionization profile [see Fig. 2]. Here, the noise causes ionization from the atomic ground state, which is different from the studied ionization of the Rydberg atoms [18]. This purely stochastic ionization for the feeble noise $\sqrt{D} = 0.00024$ is considerably smaller than the corresponding case of laser pulse with noise [Fig. 2(a)]. Hence, the observed net enhancement can be attributed to a nonlinear quantum interaction between coherent pulse and noise, which is also beyond the response to a simple quantum addition of the individual external fields.

As the noise level is further increased, we observe an enhancement of the periodic ionization profile by about 3 orders of magnitude as shown in Figs. 2(b) and 2(c). However, the increase in noise level also causes the stochastic ionization curve to rise rapidly. Eventually, for strong noise case, the coherent structures tend to wash out, and the noise dominates the ionization [Fig. 2(c)]. This suggests the existence of an optimum ratio between the noise and laser amplitudes which leads to a maximum ionization enhancement.

To quantify the quantum stochastic enhancement, we define the enhancement factor

$$\eta = \frac{P_{s+n} - P_0}{P_0} \tag{4}$$

with $P_0 = P_s + P_n$. Here, P_{s+n} denotes the average ionization probability (IP) due to the presence of the laser pulse with noise, and P_s and P_n are the individual IP for laser pulse and noise, respectively. Although this is different compared to the quantifiers commonly used [2,3], η is more suitable for our case. One can verify that a zero value of η corresponds to the case when either the laser pulse ($P_s \gg P_n$) or the noise ($P_s \ll P_n$) dominates. Furthermore, η characterizes a truly nonlinear quantum interplay as it also vanishes if we assume a "linear" response as a sum of individual IP, $P_{s+n} = P_s + P_n$. In Fig. 3, we have plotted the enhancement factor η versus the noise amplitude. It exhibits a sharp rise, followed by a maximum at a certain value of the noise (point B), and then a gradual fall off. It is worth mentioning that only a modest noise to laser ratio ($\sqrt{D_{\text{opt}}}/F_0 = 0.075$) is required to reach the optimum enhancement (here $\eta_{\max} = 36$), as indicated by a typical optimal pulse shape in the inset of Fig. 3.

The enhancement in photoionization can be understood by a simple two-step mechanism. First, starting from the ground state, the atom absorbs energy from the noise leading to an exponential population of energy levels. In a second step, the laser field causes ionization from the electron wave function exponentially distributed over many (excited) states. This simple picture is indeed verified by separating the laser irradiation and the noise input in time. A sequential application of the noise followed by the laser pulse leaves the SR curve almost invariant. On the contrary, reversing the sequence, i.e., laser pulse first and then noise, destroys the SR. Note that due to the level structure of the atom (many bound states and continuum), there is no obvious noise-induced time scale as known from two-level systems. Thus, the optimum enhancement does not show the characteristic synchronization between coherent and noise-induced time scales as in the TLS [2,3]. Indeed, in some classical systems, SR has been shown to exist without any explicit synchronization requirement [19]. Atomic ionization under a driving laser field provides the quantum analog to synchronization free SR.

It is worth mentioning that the features presented here are robust with respect to the choice of parameters. For example, we have observed the quantum SR for pulses lasting from a few cycles to few hundred cycles and for ω ranging over more than 1 order of magnitude from infrared to near UV frequencies.

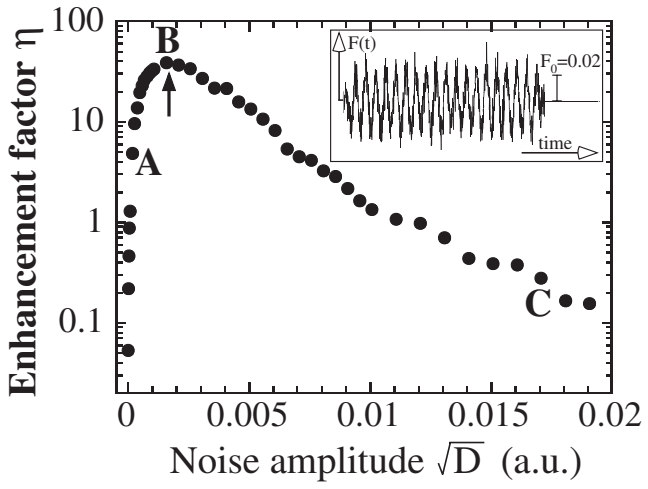

FIG. 3. The enhancement in photoionization due to quantum SR. The points marked A-C correspond to the noise amplitudes of Fig. 2(a)–2(c), respectively. Inset: A typical optimal pulse for point B with $\sqrt{D_{\text{opt}}} = 0.0015$, $F_0 = 0.02$.

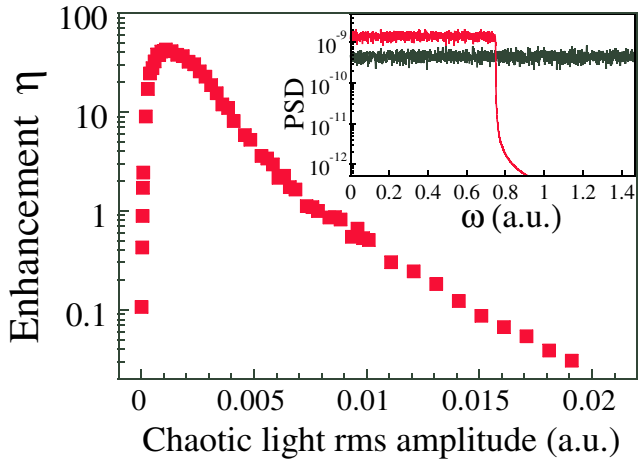

FIG. 4 (color online). Enhancement induced by a broadband chaotic light. The peak amplitude and frequency of the 20 cycle laser pulse are $F_0 = 0.02$ and $\omega = 0.057$, respectively. The bandwidth of chaotic light is $\Delta\omega = 0.75$ with central frequency $\omega_0 = 0.375$. Inset: power spectral density (PSD) of the chaotic light compared to the one for the white noise.

The intensity required for the thermal white noise renders an experimental test of our prediction quite challenging. However, one could think of replacing the white noise by a quasiwhite noise, i.e., chaotic light with a finite but broad bandwidth $\Delta\omega$. This can be realized by adding a large number (here 1024) of independent phase-randomized frequency modes. The inset of Fig. 4 shows an example of a characteristic spectral density of such a broadband source having $\Delta\omega = 20$ eV (0.75 a.u.). Modern pulse shaping techniques can generate chaotic light pulses with a bandwidth as large as 30 eV [20,21]. In Fig. 4, we have plotted η defined in Eq. (4) vs the rms amplitude of the chaotic light (analogous to the noise amplitude) for the previously used coherent pulse. One can see that such chaotic light can preserve the features of quantum SR, with an almost identical optimum compared to the one for the white noise case of Fig. 3. This observation may create new possibilities in quasicoherent control schemes of similar quantum systems.

In conclusion, we have demonstrated a new form of quantum stochastic resonance in the dynamics of the simplest atomic system for the first time. This generalized quantum SR leads to a dramatic enhancement (by several orders of magnitude) in the nonlinear ionization when a *modest* amount of optimum white noise is added to the weak few cycle laser pulse. The same effect is also achieved if one uses (realizable) broadband chaotic light instead of white noise. We emphasize that the effect is robust with respect to a range of experimentally accessible parameters. In addition to substantially broadening the existing paradigm for quantum SR to generic atomic and molecular systems, our results might provide valuable insight into the possible role of noise in designing *optimal* quasicoherent quantum control schemes [11,21]. Finally, analogous effects are also expected in other systems such as in photo-fragmentation of anharmonic diatomic molecules [22,23], and in the recently observed multiphoton transitions in current-biased Josephson devices [24], provided a coupling with an incoherent perturbation exists.

We thank A. Kenfack, W. Peijie, N. Singh, A. Buchleitner, and P. Hänggi for fruitful discussions.

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