PHYSICAL REVIEW A VOLUME 45, NUMBER 11 1 JUNE 1992

Stability of Rydberg atoms in a strong laser field

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(Received 29 July 1991)

On the basis of a classical treatment, we present analytical and numerical evidence that Rydberg atoms in a strong laser field are stable provided the orbital momentum is sufficiently large. We surmise that this effect persists in the quantum case also and we suggest its observation in laboratory experiments. The possibility that such atoms may radiate high-frequency photons is discussed.

PACS number(s): 31.50.+w, 32.80.Rm, 42.50.Hz

The investigation of the behavior of atoms under the action of a strong laser field is important for the understanding of fundamental physical phenomena concerning the radiation-matter interaction. In this connection new interesting experimental possibilities arise due to the presently available powerful sources of laser radiation [1].

Some interesting attempts to investigate these phenomena via numerical simulation have been made recently [2-4]. So far the analysis has been focused on the action of laser fields on atoms which lie in their ground state. In particular, the existence of long-living states in intense laser fields has been demonstrated [4]. In this situation, however, the field frequency is comparable to the atomic frequency (about 27 eV or 1 a.u., which we will use in the following). Such frequency is very high, and this renders experimental analysis too difficult.

A different line of investigation is concerned with the behavior of Rydberg atoms in microwave fields. The experimental work was initiated by Bayfield and Koch [5] and the theoretical analysis and understanding of this problem has been strongly connected with the manifestations of classical chaos in quantum mechanics [6,7]. Due to the high initially excited state with principal quantum number n_0, the quantum excitation process is approximately described by classical mechanics until the rescaled microwave frequency $\omega_0=\omega n_0^3$ is less than or approximately equal to 1. For $\omega_0>1$ it has been shown that quantum interference effects inhibit the classical diffusive excitation mechanism; as a consequence, the field strength required for ionization is higher than the classical one [6]. The theoretical analysis has been largely based on the reduction of the microwave ionization problem to an area-preserving map, the so-called Kepler map. It was shown [6,8] that chaotic diffusive excitation takes place only when $l<(2/\omega)^{1/3}$, where l is the orbital momentum; this is also the condition for applicability of the Kepler map. The theoretical predictions for diffusive ionization and its quantum suppression have recently been confirmed by laboratory experiments [9-11]. In such experiments with $n_0\sim 60$, $\omega_0\sim 1$, strong ionization takes place for field strengths $\epsilon\sim 10$ V/cm.

In the present paper we analyze situations in which, as we will show, it is possible to have stable, nonionized, atoms in the presence of field intensities even 6 orders of magnitude higher. In order to understand how such a possibility may arise, we recall that if the field frequency is much larger than the Kepler frequency (or the distance between two consecutive levels) then the excitation can take place only if the electron passes close to the nucleus. In the opposite case, when the electron never comes close to the nucleus, the Fourier components of the dipole moment are exponentially small. A more detailed analysis shows that exponential decay of these components takes place when the orbital momentum l satisfies the condition [6,8]

$$l>l_c\sim\left(\frac{2}{\omega}\right)^{1/3}. \tag{1}$$

For $l<l_c$, the change in the electron energy after one passage near the nucleus is described by the Kepler map and it is approximately given by $\Delta E\sim 2.6\epsilon\omega^{-2/3}$. If this quantity is larger than the ionization energy then ionization will take place after approximately one orbital period of the electron.

A completely different behavior takes place for $l>l_c$. Indeed, in this case the electron remains far from the nucleus; classical resonances do not overlap and the electron exhibits oscillations around the unperturbed Kepler orbit. This size of these oscillations is of the order of $\Delta r\sim 2\epsilon\omega^{-2}$. If Δr is less than the size of the Kepler orbit which is of order n^2, then one expects that the global motion will be stable and that the switching on of the field will not ionize the atom.

However, for very strong fields, condition (1) is not sufficient to guarantee the stability of the atom. Indeed, in such a situation, the typical value of the velocity of the electron is of the order of ϵ/ω; therefore the distance $\bar{r}$ at which the Coulomb energy becomes comparable with the kinetic energy of the electron is $\bar{r}\sim(\omega/\epsilon)^2$. It follows that the interaction time τ with the nucleus is $\tau\sim\omega\bar{r}/\epsilon\sim(\omega/\epsilon)^3$ and the change in the field's phase during this interaction is $\sim\omega^4/\epsilon^3$. As a consequence, if $\epsilon>\omega^{4/3}$, the change in this phase is small and the interaction of the electron with the nucleus is similar to a collision with an elastic ball: After one such collision, the increase of electron energy is of order $(\epsilon/\omega)^2$ and the atom will ionize [6]. From the above discussion it turns out that in order

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to have a stable atom it is necessary that the distance r between the electron and the nucleus is larger than the size Δr of the oscillations in the free field. Since $r = n^2(1-e)$, where $e = (1 - l^2/n^2)^{1/2}$ is the eccentricity of the orbit, it follows that the condition for the stability of the atom is

$$l > 2\frac{\sqrt{\epsilon}}{\omega}\left[1 - \frac{\epsilon}{\omega^2 n_0^2}\right]^{1/2}. \qquad (2)$$

Therefore we expect the atom to be stable if the initial orbital momentum l is larger than the maximum of the two values given by (1) and (2). Of course, a necessary condition is $2\epsilon/\omega^2 < n_0^2$ or $\epsilon_0 < \omega_0^2/2$, where $\omega_0 = \omega n_0^3$ and $\epsilon_0 = \epsilon n_0^4$ are the usual rescaled field frequency and intensity, respectively.

In order to check the above predictions we integrated the classical equations of motion for a hydrogen atom in a linearly polarized electric field. We restrict ourselves to states with zero value of the projection of the orbital angular momentum along the field direction (magnetic quantum number $m=0$). Due to the symmetry of the Hamiltonian this projection is an integral of the motion and the orbits of the electron lie in a plane. The Hamiltonian has the form

$$H = \frac{p_x^2}{2} + \frac{p_y^2}{2} - \frac{1}{(x^2+y^2)^{1/2}} + \epsilon x \cos\omega t \, . \qquad (3)$$

We have chosen an initial distribution of trajectories corresponding to fixed principal and orbital quantum numbers, n_0 and l, and with uniform distribution in the conjugated phases, λ and ψ. We organized the computer code in such a way as to have efficient computations in both opposite cases when the electron is close or far from the nucleus. The switching on (and off) of the field ϵ has a sine-square shape ($\sin^2\omega t/N$) and takes place in a number of periods of the external field $N=50$. However, we checked that changing the switching time N does not appreciably influence the results.

A characteristic feature of the motion are the fast oscillations in the strong laser field superimposed on the slow motion along the Kepler orbit. A typical example is shown in Fig. 1(a). Since the size of the oscillations is less than the distance from the nucleus, the strong laser field does not lead to ionization. In Fig. 1(a) we show only a few periods of the orbit; however, we checked the stability of the atom up to a thousand orbital periods of the electron. In the opposite case, when the size of the oscillations is comparable to the distance from the nucleus, after a few orbital periods the electron collides with the nucleus and the atom ionizes [Fig. 1(b)].

In Fig. 2 we plot the survival probability W_s (the fraction of nonionized atoms) as a function of the field strength ϵ_0 for fixed initial $l/n = 3/4$ and fixed frequency $\omega_0 = 50$. The survival probability W_s is equal to unity up to a quite large critical value $\bar{\epsilon}_0$. This means that the atom remains stable even in a very strong field provided that the orbital momentum is large enough. We may compare this critical value $\bar{\epsilon}_0$ with the ionization border (which coincides with the chaos border) $\epsilon_c = 1/50\omega_0^{1/3}$, which is correct for trajectories with a small value of angular momentum ($l < l_c$). The ratio of the two critical

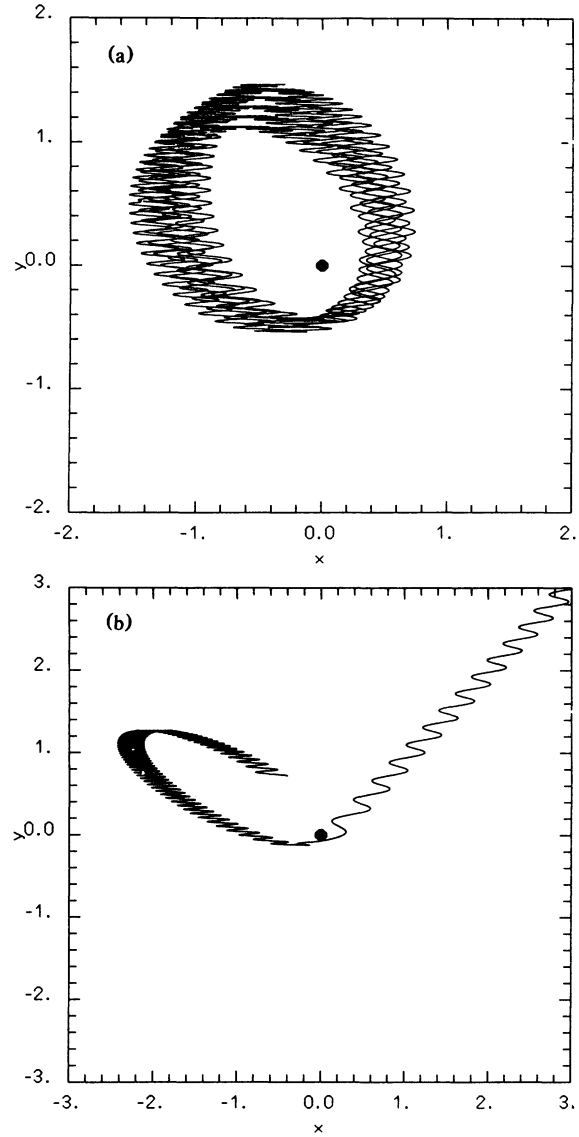

FIG. 1. (a) A classical trajectory for a stable Rydberg atom in a laser field with $\epsilon_0 = 400$, $\omega_0 = 50$; the eccentricity of the orbit is $e \approx 0.5 (l/n \approx 0.85)$. 250 field periods are shown. (b) An ionized classical trajectory for the same field strength ϵ_0 and frequency ω_0 as for (a). The eccentricity is $\epsilon \approx 0.9 (l/n \approx 0.45)$.

field values for ionization gives $\bar{\epsilon}_0/\epsilon_c \sim 10^5$.

For the case of Fig. 2 the analytical estimate (2) gives $\bar{\epsilon}_0 \approx 420$ in agreement with numerical results. For $\epsilon_0 < \bar{\epsilon}_0$ the energy of the electron after the laser pulse was near its initial value. For $\epsilon_0 > \bar{\epsilon}_0$ practically all trajectories ionize and the transition around $\bar{\epsilon}_0$ is quite sharp. Moreover, for almost all trajectories, ionization occurs after only 1–3 passages near the nucleus corresponding approximately to 50–150 field periods. We checked that the remaining very few trajectories which for $\epsilon_0 \gg \bar{\epsilon}_0$ are not ionized during the interaction time are due to the fact that after one passage near the nucleus the electron energy becomes very close to the ionization border, so that the orbital period at that energy is larger than the interaction time.

We also checked that this survival probability remains

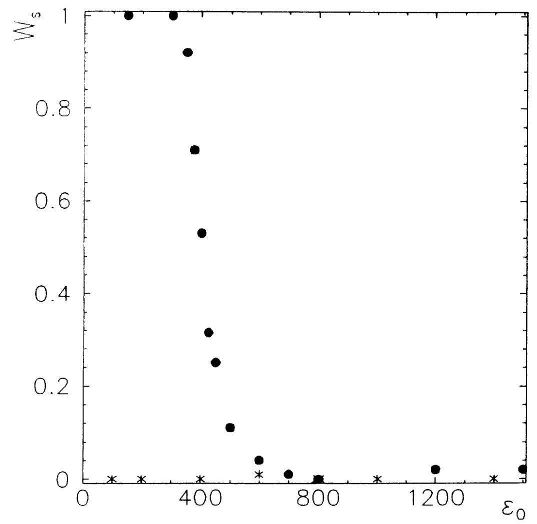

FIG. 2. The fraction of nonionized atoms W_s vs the field strength ϵ_0 for $\omega_0=50$. Solid circles correspond to initial orbital momentum $l/n=0.75$; asterisks correspond to $l/n=0.8$. The interaction time with the field is equal to 5000 field periods, and the switching on and off of the field is equal to 50 field periods.

at the same level with increasing field intensity up to $\epsilon_0 \approx 25\,000$. Therefore we may conclude that above the critical border (2) the atom is completely destroyed after one or two Kepler periods.

We would like to stress the important difference between our results and previously published papers [1–4,12–16]. In our case there is no atom stabilization: Above the ionization border (2), which is quite high, the atom is destroyed. However, our considerations were carried out for the classical atom and the conditions for their applicability to real atoms are $n_0 \gg 1$ and $n_0^{-3} \ll \omega \ll 1$. Due to this fact our results do not contradict the numerically observed stabilization for the hydrogen atom with $\omega \approx 1$.

The asterisks in Fig. 2 give the survival probability W_s for the case $l/n_0=0.3<l_c/n_0$. Since stability conditions (1) and (2) are violated, ionization takes place for all values of ϵ_0, down to the chaos border $\epsilon_0 \sim 0.1$.

The above numerical results support the theoretical prediction that stable states in strong fields can exist only for relatively large values of orbital momentum.

The properties of the motion in the case of stable states can be understood on the basis of an averaging procedure over the fast oscillations of the electron. To this end it is convenient to move to another reference frame in which no field acts on the electron, and the nucleus oscillates according to the law $(\epsilon/\omega^2)\cos\omega t$ [1,6]. In this frame the potential is

$$U=\frac{1}{\{[x-(\epsilon/\omega^2)\cos\omega t]^2+y^2\}^{1/2}}\,. \tag{4}$$

Under the assumption that the amplitude of these oscillations is small and averaging over the fast oscillations (like in the case of the Kapitza pendulum), one obtains the effective Hamiltonian $H_{\rm eff}$, which describes the average motion of the electron far from the nucleus,

$$H_{\rm eff}=\frac{p_x^2}{2}+\frac{p_y^2}{2}-\frac{1}{r}+\frac{\epsilon^2}{4\omega^4 r^5}(r^2-3x^2)+\frac{\epsilon^3}{4\omega^6 r^9}[(r^2-3x^2)^2+9x^2y^2]\,. \tag{5}$$

Since $\omega_0 \gg 1$, the last term in Eq. (5) can be neglected. The preceding one is small in comparison with the usual Coulomb potential and mainly leads to a regular precession of the ellipse with fixed angular velocity and fixed value of the orbital momentum (the problem is equivalent to the motion of a *sputnik* in the field of a compressed earth) [17]. The frequency of precession is equal to $\omega_{\rm pr}=3\epsilon^2/8\omega^4 n^3 l^4$. Our numerical measurement of the precession rate for the averaged motion were in agreement with this value.

As to the possibility of laboratory experiments, a convenient choice seems to be a CO_2 laser with $\omega \approx 0.1$ eV $\approx 3.7\times10^{-3}$ a.u. With such frequency, the case of Fig. 2, where $\omega_0=50$, corresponds to atoms with $n_0=24$. According to the data of Fig. 2 the atoms will remain stable up to a field intensity $\bar{\epsilon}=\bar{\epsilon}_0/n_0^4 \approx 6\times10^6$ V/cm. Another convenient case for the experimental observation of the effect predicted in this paper could be, for example, $n_0=50$; for a CO_2 laser $\omega_0 \approx 450$, and the atoms with orbital quantum number $l \approx 30$ will be stable up to a field intensity $\epsilon \approx 3\times10^4/n_0^4 \approx 2.5\times10^7$ V/cm. Since there is a well-developed technique to create Rydberg atoms [9–11], the observation in laboratory experiments of the stable states predicted here should be quite possible. At first glance, the existence of such stable states in strong laser fields appears to be quite unexpected since the energy of photons is larger than the ionization energy. However, it turns out that for large values of orbital momentum the matrix elements for coupling with the continuum become very small. This fact can be understood on classical grounds; indeed, the matrix elements are determined by the Fourier components of the classical motion which are very small for classical stable states where the electron moves far from the nucleus.

An interesting consequence of the existence of stable atoms in strong field is the fact that there is a large shift of energy ($\Delta E \approx \epsilon^2\omega^{-2}/2$) for high principal quantum numbers, while there is a practically no shift for the ground state of the atom, for which the field acts as a small perturbation (ϵ_0 and $\omega_0 \ll 1$). For the case considered above $n_0=50$, the difference in energy is near 34 eV. In principle, the transition from the excited state to the ground state can take place and one can observe photons with this energy. A possibility to have relatively high probability of such transitions is to have stable atoms which interact with the laser field during a long time. Another possibility is to increase the intensity of the laser field in such a way that the electron will pass near the nucleus, where the probability of being captured in the ground state is relatively high.

Note added. After submission of this paper a new interesting paper was published [18] in which, on the base of quantum computations, stabilization was found for $\omega \ll 1$. We would like to stress that this is not in contradiction with our classical results, since in our case $m=0$ while in [18] $m=5$. Moreover, our present computations show that the value of m is of crucial importance for the stabilization problem.

One of us (D.S.) would like to thank University of Milano for the hospitality during the period when this work was performed and N. B. Delone for stimulating discussions. This work was performed with the support of Consiglio Nazionale delle Ricerche, Italy.

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