New States of Hydrogen in a Circularly Polarized Electromagnetic Field

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We predict the existence in hydrogen of stable nonspreading Rydberg wave packet states that have no classical counterparts. We show their time-dependent creation from circular Rydberg states in numerical simulations. These states are both angularly and radially localized exactly in antiphase from the corresponding nonspreading (so-called Trojan) packet position and are stabilized by quantum interference. [S0031-9007(96)01189-1]

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The description of fundamental quantum systems in regimes of strong perturbation is a matter of wide interest in physics. One-electron bound systems are the most fundamental at the atomic level, and striking new phenomena have been predicted and observed in the past decade or so in such systems. These have come from experiments in laser fields [1] or in static electric and magnetic fields [2], in experiments on field-free wave packets at the quantum-classical interface [3], and experiments with time-dependent microwave fields [4]. We are interested here particularly in resonance phenomena at field strengths near to one scaled atomic unit, $\mathcal{E}_{\rm at} = 1/n^4$. Such values can be achieved with very high power lasers or, after excitation to high values of the principal quantum number n, with modest laboratory fields.

In this paper we report the identification of a new type of state in strongly perturbed hydrogen. By applying an approximate, analytic, and nonperturbative "quantum pendulum" description to hydrogen in a strong scaled radiation field, we have previously provided [5] the quantum theory of a family of classically stable wave packets in a circularly polarized (CP) radiation field [6]. These have been called "Trojan packets" because of their analogy to the Trojan asteroids observed in the solar system. Any quantum packet exhibiting the tendency to evolve without spreading is an anomaly in quantum theory, and features of Trojan packets have been the subject of interesting reports recently [7], including relatively high precision estimates of their very great ionization lifetime [8].

We now report for strong-field hydrogen the identification of a new set of states, which are classically unstable and therefore without any celestial analogs, but are quantum stabilized. The new states can be called "anti-Trojan" states because the theory indicates that an anti-Trojan twin should be found exactly opposite each Trojan on its orbit. According to extensive numerical simulations that we have made to confirm our analytic theory, these states appear to be relatively robust and accessible to long-term experimental study. Both the method of dynamical formation of the classically unstable anti-Trojan state and its character as a new type of quantum state of strong-field hydrogen are interesting.

In order to present the anti-Trojan states most simply we first summarize our quantum pendulum theory. The hydrogenic Hamiltonian in the time-dependent CP field becomes time independent in a frame rotating at the CP frequency ω and is given by

$$H_{\rm CP} = \frac{\mathbf{p}^2}{2} - \frac{1}{|\mathbf{r}|} + \mathcal{E}x - \omega L_z \,. \tag{1}$$

The exact time-dependent and fully field-dressed states of hydrogen are obtainable from the eigenstates of this static rotating-frame Hamiltonian, if we can solve the time-independent eigenvalue problem $H_{\rm CP}\Phi = E(\mathcal{E})\Phi$.

Of course, $H_{\rm CP}$ is characterized by a conflict between cylindrical and spherical symmetries. However, we have already shown [5,9] the following. If Φ is expanded in the bare hydrogenic basis, $\Phi = \Sigma_{n,l,m} c_{nlm} R_{nl} Y_{lm}$, then the energy eigenvalue problem for $H_{\rm CP}$ can be solved by block diagonalization following a suitable rearrangement of the n, l, m summation. The approximations involved are nonperturbative and have been tested for Φ's that represent localized and near-circular packet states.

The blocks into which the bare matrix elements of $H_{\rm CP}$ must be grouped are most easily organized by inverting the order of "importance" of the hydrogenic quantum numbers. Within a given block labeled by λ and ν the differences $\lambda = l - m$ and $\nu = n - l - 1$ are fixed. The next step is to concentrate attention on a high value of m with the helicity of the CP field, so $m \equiv m_0 + \mu$ ($m_0 \gg 1$). Pure circular states have therefore the quantum numbers $\{\mu\lambda\nu\} = \{\mu 00\}$.

The bare matrix elements of $H_{\rm CP}$ are next labeled by μ, λ, ν in the circularity sequence just described. For given λ and ν, the variation of μ outlines a block, which is necessarily infinite in size. Since n, l, and m are rigidly linked in any block, the block space is one dimensional and μ is the running parameter. To justify the nuisance of this block reorganization, one must verify that the blocks are independent. Of course this is not strictly true, and this is the principal approximation of

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the method. We are most interested in states relatively well localized in all three dimensions, with small values of ν and λ. This permits the approximation in which dipole matrix elements within a block are set equal, with the common value $r_0/2 \equiv m_0^2/2$, and matrix elements between blocks are set equal to zero. The expansion of the bare eigenvalue $E_n = -(2n^2)^{-1}$ around $m_0 + \lambda + \nu + 1$ up to second order in μ, and a choice of m_0 that conforms to Keplerian resonance with the CP field: $\omega = 1/(m_0 + 1)^3$ leads to a simple three-term recursion for the wave function expansion coefficients,

$$\frac{\mathcal{E}}{2} r_0[A^{\lambda\nu}_{\mu+1} + A^{\lambda\nu}_{\mu-1}] = \Big[E(\mathcal{E}) + \frac{1}{2(m_0 + \lambda + \nu + 1)^2} + m_0\omega + \frac{3\mu^2}{2r_0^2} \Big] A^{\lambda\nu}_{\mu} . \quad (2)$$

Here the A's are the c's with indices shifted by the block reorganization.

This recursion relation implies a differential equation for the generating function $\psi_{\lambda\nu} \equiv \Sigma_\mu A^{\lambda\nu}_\mu \exp[i\mu\phi]$, and this equation is just an energy eigenvalue equation $\mathcal{H}\psi_{\lambda\nu} = \kappa^j(\mathcal{E})\psi_{\lambda\nu}$, where $\mathcal{H}$ is the effective Hamiltonian

$$\mathcal{H} = \frac{3}{2}\frac{1}{r_0^2}\frac{\partial^2}{\partial\phi^2} + \mathcal{E} r_0 \cos\phi , \quad (3)$$

$r_0 = m_0^2$, and $\mathcal{E}$ has been chosen positive. The effective Hamiltonian (3) can be recognized as describing a quantum pendulum (with *negative* scaled mass $= -1/3$, and effective gravitational constant $g = 3\mathcal{E}$). For simplicity of comparison with reference works [10], the redefinition of parameters according to $\alpha^j \equiv -(8r_0^2/3)\kappa^j(\mathcal{E})$, $2\xi \equiv \phi - \pi$, and $p \equiv 4\mathcal{E}/3\omega^2$ converts the pendulum equation into Mathieu's equation in standard form.

The field-dressed eigenvalues $E(\mathcal{E})$ of $H_{\rm CP}$ depend on the Mathieu index j, as well as on λ and ν, and are given by

$$E^j_{\lambda\nu}(\mathcal{E}) = -\frac{1}{2(m_0 + \lambda + \nu + 1)^2} - m_0\omega + \kappa^j(\mathcal{E}) . \quad (4)$$

We now apply the results just obtained and plot the energies κ^j. Figure 1 shows the predicted energy spectrum as a function of the external field parameter $p = 4\mathcal{E}/3\omega^2$. [In dimensional units this parameter is just $4/3$, the ratio of the radius of the orbit of a free electron in the presence of a CP radiation field (pure Volkov problem) to the Bohr radius (pure Coulomb problem).]

For the quantum pendulum the localization of the wave function occurs around the two points $\phi = 0$ and $\phi = \pi$. The first point of state localization is the Trojan stable point at $\mathbf{r} = [m_0^2, \phi = 0]$, and localization there corresponds to the *highest* excited state of the m_0 Hamiltonian $\mathcal{H}$ (recall the negative effective mass) as was defined first in harmonic approximation [6] and more generally in [5].

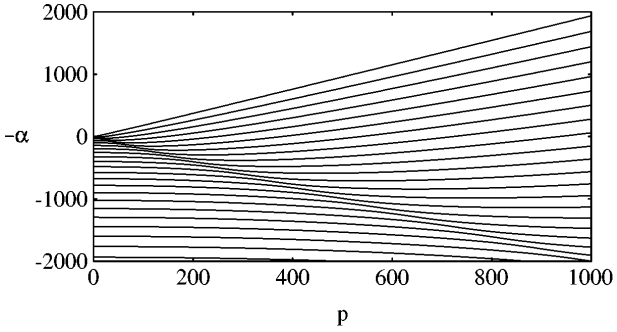

FIG. 1. Levels of scaled energy $-\alpha = (8r_0^2/3)\kappa$ (corresponding to symmetric Mathieu functions) as functions of the parameter p. The value $-\alpha = 2p$ corresponds to Trojan packets. States with scaled energy in the vicinity of the line $-\alpha = -2p$ are anti-Trojan packets. Note that anti-Trojan packets do not belong to a single energy line but to different lines close to the spectrum folding around $\alpha = 2p$.

The second localization point is of a different character because it corresponds to a packet state that is both 3D localized and nonspreading, and has no classically stable counterpart. It is predicted by our pendulum theory to be located opposite the first point, at $\mathbf{r} = [m_0^2, \phi = \pi]$. State localization around the second point is purely quantum mechanical in character and is an example of the quantum scar effect, known from the properties of quantum systems that are classically chaotic [11]. The present analysis of 3D anti-Trojan states also provides a theory for recent 1D numerical observations of Buchleitner and Delande [12].

What are the criteria that can be used here to identify state localization around the unstable point? We first look for a local maximum in the density of states. This is an imprint of localization of the eigenstate with the corresponding energy, around the trajectory in question [11]. As Fig. 1 shows, there is a fold or kink around the energy line $\kappa = -r_0\mathcal{E}$ ($\alpha = 2p$). This corresponds to the separatrix in pendulum phase space and shows a larger density of levels in the vicinity of this energy. This nonclassical localization can also be expected from the properties of an asymmetric rotor, which can be mapped onto a similar quantum pendulum [13]. Our partly heuristic arguments can also be checked directly from the properties of the symmetric Mathieu function $e_{2j}(\xi)$ [10,14].

The interval of field strengths for which the Trojan (stable-point-related) packet exists has been expressed in terms of the scaled field $\mathcal{E}_{\rm sc} = \mathcal{E}\omega^{-4/3}$ as $\mathcal{E}_{\rm sc} < 2/9^{4/3} \approx 0.1068$ [6]. A more interesting condition can be obtained for anti-Trojan packets. We have found, for sufficiently large m_0,

$$\mathcal{E}_{\rm sc} < \mathcal{E}_{\rm cr} \equiv 3/4m_0 . \quad (5)$$

In contrast to the condition for the existence of Trojan packets, the anti-Trojan critical field $\mathcal{E}_{\rm cr}$ is not independent of m_0. In particular, the critical field vanishes when

$m_0 \rightarrow \infty$, which is consistent with the purely quantum-mechanical nature of anti-Trojan wave packets.

In order to test our theory we have also solved numerically the time-dependent and field-dependent Schrödinger equation for the electron wave function. For this test, in order to step as far away from our pendulum theory as possible, we used a time-dependent fast Fourier transform method on a spatial grid [15], rather than an expansion in bare-state amplitudes. The coordinate along the CP propagation direction is nonrelativistically absolutely stable so we ignored it, under both Trojan and anti-Trojan conditions. We started from a prepared circular state and turned on the CP field in a quasiadiabatic manner for 20 CP field cycles and then held the CP amplitude constant at the value $\mathcal{E}_0$ for an additional 10 cycles. For computational convenience, the frequency of the field was chosen equal to the Kepler frequency of the $m_0 = 19$ circular state.

In the Trojan case, shown here for comparison, we took $\mathcal{E}_0 = 0.016\omega^{-4/3}$. Figure 2(a) shows contour plots of the probability density calculated from the resulting wave function. Note that for the chosen phase of the field the point of electron localization at the end of each cycle lies approximately at the right side of the nucleus.

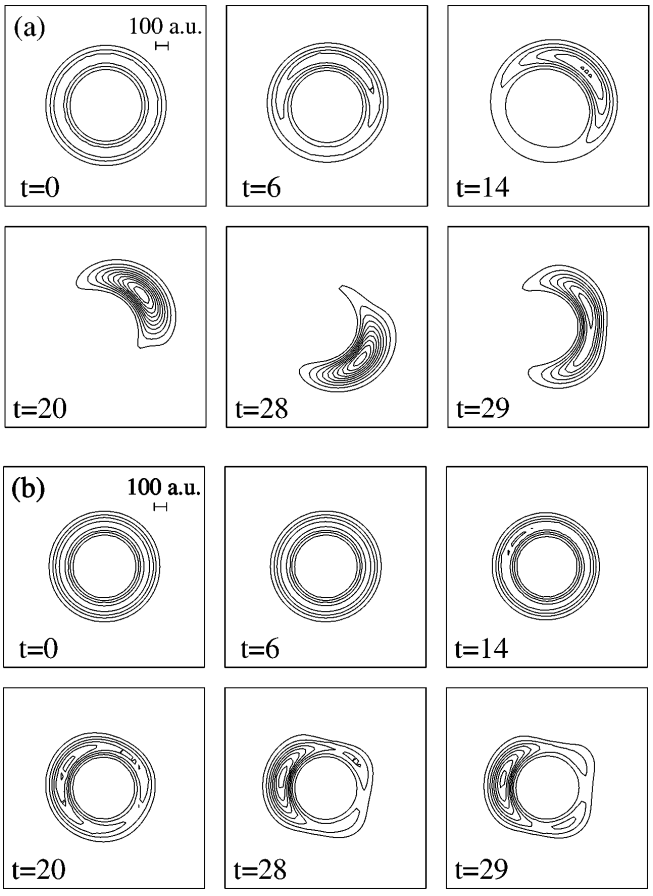

FIG. 2. (a) Trojan wave packet formation. The initial state is a circular state with $m_0 = 19$. (b) Anti-Trojan wave packet formation. The initial state is a circular state with $m_0 = 18$. Note that localization occurs at the opposite side of the nucleus from the Trojan case. In both (a) and (b) the CP field was switched on quasiadiabatically, and $\omega = 1/20^3$; the small horizontal bar sets the spatial scale. (Electron probability density contours are plotted on a linear scale, and the time units are CP field cycles.)

In contrast to Trojan wave functions, anti-Trojan wave functions converge in the zero-field limit ($p \rightarrow 0$) to hydrogenic eigenstates for which the resonant condition $\omega = 1/n^3$ is not satisfied exactly. This is caused by the nonlinearity of the Coulomb spectrum, represented within our pendulum model by the kinetic energy term in the Hamiltonian (3). Therefore, in the anti-Trojan case, we applied a field with the same frequency and $\mathcal{E}_0 = 0.0064\omega^{-4/3}$, but the initial state was the circular state with magnetic quantum number one atomic unit lower, i.e., $m_0 = 18$.

The field in this case also generates a packet state, shown in Fig. 2(b), which orbits around the nucleus, but the point of electron localization is shifted approximately 180° with respect to the previous case so we observe the probability density concentrated at the left side of the nucleus at the end of each cycle. Because of its purely quantum-mechanical stability, the anti-Trojan packet requires some weak "wings" that reach to each other around the nucleus, resulting in a characteristic "pull-tab" shape.

To evaluate these observations quantitatively we have calculated a localization function $R(t)$, which is the electron probability within a fixed rectangle through which the packets sweep in their orbits (Fig. 3). Periodic oscillations of $R(t)$ show that well-localized packets are

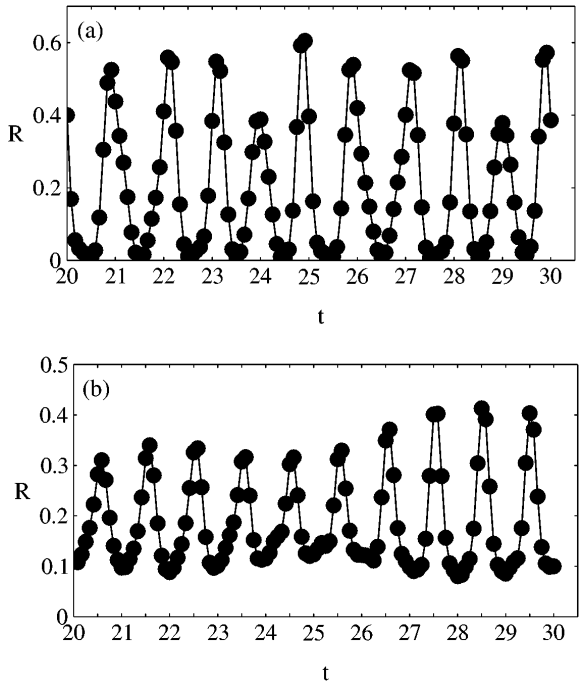

FIG. 3. Localization functions $R(t)$ as described in the text, and calculated from the states plotted in Figs. 2(a) and 2(b) after the fields have reached their steady values. (Time in CP field cycles.)

generated. Note that the maxima of the function $R(t)$ for the Trojan state are shifted almost exactly half a cycle with respect to the corresponding maxima for the anti-Trojan state. In the anti-Trojan case this function shows 75% excess of probability on one side of the nucleus with respect to the other, which can be considered a fairly high standard of localization for the relatively low parameter $n = 20$ used in our simulations. Details of additional quantitative checks will be given elsewhere.

In summary, we have described a new class of nonspreading packet states that are simultaneously radially and angularly localized and which, in contrast to Trojan packet states, are classically nonstable and are stabilized by quantum interference around the orbit. We have confirmed the validity of our pendulum theory by independent numerical calculations. As for physical realizability, the anti-Trojans are predicted to arise when parameter values are consistent with $\mathcal{E} \approx 1000$ V/m for $m_0 = 50$, comfortably within the range of current microwave experiments on Rydberg systems.

There are a number of directions in which we expect the theory we have presented here to be applied. We note that the rotating frame CP Hamiltonian is the same as the Hamiltonian for hydrogen in static electric and magnetic fields together, if just the paramagnetic term is retained. The theory suggests simple methods for creating nonfield-dependent wave packets that are nonetheless well localized both radially and angularly. The approximation of near circularity that motivates our block diagonalization is a kind of rotating wave approximation and so has obvious relevance to linear polarization. Of course, our entire procedure is, in a loose sense, an extension of the method for assembling a quasiclassical wave packet outlined so clearly by Brown [16]. As might be expected, experience with our theory also allows insights into other atomic packet phenomena, but these will have to be presented elsewhere.

Note added—An interesting related paper from Brunello, Uzer, and Farrelly came to our attention after submission [17].

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