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Ionization of Rydberg atoms by circularly and elliptically polarized microwave fields

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A classical study of the dynamics of atomic Rydberg states interacting with both elliptically and circularly polarized microwave fields is presented. In recent experiments, enhanced ionization rates were observed in elliptically as opposed to circularly polarized fields. A classical phase-space simulation provides good qualitative agreement with experimental findings. These results are explained in terms of the breakdown of almost conserved quantities as the polarization of the field is changed from circular to elliptical.

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In a recent experiment Fu *et al.* [1] made the first observations of the ionization of (sodium) Rydberg atoms by circularly (CP) and elliptically polarized (EP) (8.5-GHz) microwave fields. These experiments revealed a substantially higher threshold field for ionization in the case of circular polarization than that for a linearly polarized (LP) or EP field. Fu *et al.* [1] explained their CP results by going to a rotating frame and proposing that in this (time-independent) picture ionization proceeds in essentially the same way as for a static field. Subsequently, Nauenberg [2] argued that the ionization mechanism in CP fields was considerably more complicated than suggested by Fu *et al.* [1] and that the effect of rotation on the ionization threshold must be included. In this Rapid Communication we present a classical picture of ionization of the hydrogen atom by elliptically and circularly polarized fields which provides a good account of the differences in ionization rates observed experimentally. Of special importance is the EP case, which is a nontrivial intermediate case between the CP and LP limits.

In Cartesian coordinates and atomic units and assuming the dipole approximation to be valid, the Hamiltonian for a hydrogen atom subjected to a time-dependent microwave field (field strength F) of arbitrary polarization can be written as follows,

$$H=\tfrac{1}{2}(P_x^2+P_y^2+P_z^2)-1/r+F[x\cos\omega t+y\cos(\omega t+\alpha)]\,, \tag{1}$$

where α is a phase factor; $\alpha=0(\pm\pi/2)$ corresponds to linear (circular) polarization. Intermediate values correspond to elliptical polarization. In the limit $\omega=0$ the system is simply the linear Stark Hamiltonian. In the CP limit this Hamiltonian arises in a variety of guises; it has been studied as a model for the effect of radiation pressure on the dynamics of dust particles orbiting a planet [3] and also in the context of circular dichroism [4].

In the CP limit, transformation to a rotating (at angular velocity ω) Cartesian coordinate system (u,v,z) yields the Hamiltonian

$$H=\tfrac{1}{2}(P_u^2+P_v^2+P_z^2)-1/r-\omega L_z+Fu=H_0+H_1\,, \tag{2}$$

where

$$L_z=uP_v-vP_u \tag{3}$$

and H_1 contains the terms in F and ω. In the rotating frame the problem is equivalent to that of a hydrogen atom subjected to crossed electric and magnetic fields. The Hamiltonian (2) possesses a number of interesting features: If a particle is started out in the plane of polarization ($z=0$) with $P_z=0$ then it remains in that plane permanently, allowing a reduction in dimensionality to be achieved. This special case has been the subject of several theoretical and numerical studies [2,5,6]. Although the problem resembles the dc Stark effect the Hamiltonian is nonintegrable because L_z is no longer conserved. Averaging of the perturbation is straightforward and simply involves making the Pauli replacement. The averaged Hamiltonian is the following,

$$H=\tfrac{1}{2}(P_u^2+P_v^2+P_z^2)-1/r-\omega L_z-\tfrac{3}{2}nFA_u=H_0+H_1'\,, \tag{4}$$

where A_u is the component of the Runge-Lenz vector in the u direction in the rotating frame. This Hamiltonian represents a separable approximation to the original system as can easily be seen by constructing two angular momenta from **L** (orbital angular momentum) and **A** (Runge-Lenz vector):

$$\mathbf{M}=\tfrac{1}{2}(\mathbf{L}+\mathbf{A}),\quad \mathbf{N}=\tfrac{1}{2}(\mathbf{L}-\mathbf{A})\,. \tag{5}$$

This allows the averaged perturbation to be expressed in terms of two uncoupled spins,

$$H_1'=-\omega(M_3+N_3)-\tfrac{3}{2}nF(M_1-N_1)\,. \tag{6}$$

H_1' can also be written in terms of action-angle variables

by using the results [7]

$$M_1=(M^2-M_3^2)^{1/2}\sin\phi_M,\quad N_1=(N^2-N_3^2)^{1/2}\sin\phi_N,$$
$$M_2=(M^2-M_3^2)^{1/2}\cos\phi_M,\quad N_2=(N^2-N_3^2)^{1/2}\cos\phi_N, \tag{7}$$

where M_3 and N_3 are now thought of as classical actions. An even simpler representation is possible by noting that $\lambda(A_u,A_v,L_z)$ constitutes an angular momentum associated with the symmetry chain $SO(4)\supset SO(3)_\lambda\supset SO(2)$. The perturbation is a linear combination of two components of λ and an SO(4) rotation thus allows all angle dependence in the Hamiltonian to be removed. For present purposes this is less useful than the action-angle representation of Eq. (7) because the rotation needed depends on the frequency and strength of the field. In the case of an EP field it is still possible to go to a rotating frame as before except that the transformation does not remove the time dependence. Instead, the perturbation contains driving terms at twice the actual field frequency. The rotating frame remains a useful device, however, if one is interested in understanding the effect of deviations from circular polarization on the dynamics.

The classical dynamics was examined by integrating Hamilton's equations of motion in the rotating frame for a field of specified polarization and $\omega=8.5$ GHz [1]. In order to avoid singularities associated with the Coulomb term in the potential the Kustaanheimo-Stiefel (KS) transformation was used to regularize the motion [8]. To deal with the explicit time dependence (in the case of LP or EP fields) the Hamiltonian was first transformed into an extended phase space after which the KS transformation resulted in a five-dimensional (5D) time-independent Hamiltonian. The KS procedure has the advantage that the connection with the angular momenta in Eq. (6) can be made readily and the integration is extremely stable numerically. The scaling property of the problem in the dipole approximation, $\omega\rightarrow\kappa\omega$, $E\rightarrow\kappa^{2/3}E$, and $F\rightarrow\kappa^{4/3}F$, was used in the actual integrations. Classical phase-space averaging was used to determine ionization probabilities in which a microcanonical ensemble of 300 randomly chosen initial conditions (at each polarization) was integrated for 50 periods using a trapezoidal turn-on and turn-off of the field [9]. The turn-on and turn-off times were each 5% of the total integration time. A major difference between this study and the well-studied LP case (for example, Grochmalicki, Lewenstein, and Rzazkewski [10] who studied stabilization in an intense LP laser field) is that the Hamiltonian is three-dimensional. In the LP case one can take advantage of conservation of m and obtain a 2D Hamiltonian (in cylindrical coordinates). This provides a substantial reduction in computational effort as compared to the 3D case. Ionization probability as a function of polarization is shown in Fig. 1 and the results are in good qualitative agreement with experimental results presented for Na in Ref. 1. Specifically, the rapid decrease in ionization probability as the CP limit is achieved is apparent in Fig. 1 and should be compared to the experimental findings in Ref. [1].

The 3D nature of the system makes the construction of Poincaré surfaces of section impractical; an exception is

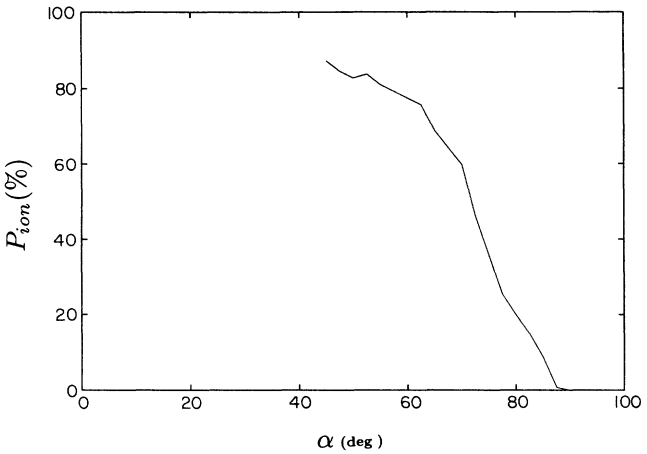

FIG. 1. Percentage ionization (P_{ion}) obtained by a classical phase-space calculation as a function of polarization angle α. The data is for $n=46$, $F=110$ V cm^{-1}, and $\omega=8.5$ GHz between $\alpha=45°$ and 90°, the latter corresponding to circular polarization of the field.

for motion confined to the plane of polarization in a CP field. An indication of the structure of phase space can be obtained by comparing the exact dynamics to that predicted by the averaged Hamiltonian (6). In accord with the observations of Nauenberg [2] in the CP case examination of surfaces of section reveals that relatively little of the phase space is chaotic below the classical ionization threshold, $F\approx 1/10n^4$ for motion in the plane of polarization [5,6]. To a good approximation the dynamics is described by the spin Hamiltonian of Eq. (6). This is also in accord with the quantum calculations of Fu *et al.* [1] for hydrogen which revealed extremely small avoided crossings between different n manifolds for the hydrogen atom. The CP case is unusual in that although there are no exact constants of motion besides the energy (in the rotating frame), the problem is not only almost integrable but almost separable also.

An EP field introduces time-dependent terms into the Hamiltonian (in the rotating frame) which effectively rules out the construction of Poincaré surfaces of section. Even for motion restricted to the plane of polarization, phase space is 5D so "double" surfaces of section are of little use and, in principle, Arnold diffusion can occur. The approximate separability of the problem in the CP limit suggests investigating a similar approximation in the EP case. Making the Pauli replacement twice in Eq. (1) gives

$$H=\tfrac{1}{2}(P_x^2+P_y^2+P_z^2)-1/r$$
$$-\tfrac{3}{2}nF[A_x\cos\omega t+A_y\cos(\omega t+\alpha)]. \tag{8}$$

Within a given n manifold and in terms of the angular momenta M and N the Hamiltonian becomes

$$H=\frac{-1}{2n^2}-\frac{3}{2}nF[(M_1-N_1)\cos\omega t$$
$$+(M_2-N_2)\cos(\omega t+\alpha)] \tag{9}$$

and is equivalent to the twin spin systems

$$H_M = -\tfrac{3}{2} nF[M_1 \cos\omega t + M_2 \cos(\omega t + \alpha)] ,$$
$$H_N = \tfrac{3}{2} nF[N_1 \cos\omega t + N_2 \cos(\omega t + \alpha)] . \quad (10)$$

In the opposite limits of LP and CP fields these Hamiltonians are integrable. The conserved quantities in these limits are $H_M - \omega M_3$ and $M_\omega = M_1 + M_2$, respectively, for H_M with similar expressions for H_N. M_ω is the component of M along the field direction and corresponds to conservation of m in the LP limit. In the EP case numerical construction of surfaces of section reveals that the Hamiltonians (10) are also integrable. Averaging the perturbation in this way is clearly too drastic to provide any insight into the enhancement of ionization rates as the polarization is changed, since all chaos within a given n-manifold is suppressed and diffusion in principal action space is prohibited. In essence, this approximation is akin to the rotating-wave approximation [6].

Further insight into the ionization dynamics in the EP case may be obtained by making a different approximation. The discussion is restricted to motion in the plane of polarization, and cylindrical coordinates in the rotating frame will be used. (Equivalent results are obtained by working in the original nonrotating frame.) The following Hamiltonian is obtained,

$$H = \frac{1}{2}\left[P_\rho^2 + \frac{P_\phi^2}{\rho^2}\right] + \omega P_\phi - \frac{1}{\rho} + V(F,\omega,\alpha;\rho,\phi,t) , \quad (11)$$

where P_ϕ corresponds to L_z. As noted, this Hamiltonian does not permit the construction of surfaces of section as it stands because the effective phase space is 5D. However, an adiabatic separation of the ρ and ϕ motion is possible in a CP field provided the frequencies in the ρ and ϕ motion are sufficiently different. This kind of adiabatic separation is well known in nonlinear classical dynamics, and is similar to the reduction of the LP Hamiltonian to 1D in parabolic coordinates [11]. In the present case the adiabatic approximation will be valid in the limit of circular Rydberg states where $m \approx n-1$ [12]. Generalizing to the EP case gives an effective Hamiltonian,

$$H' = \frac{P_\phi^2}{2\bar{\rho}^2} + \omega P_\phi + V(F,\omega,\alpha,\bar{\rho};\phi,t) , \quad (12)$$

where ρ now appears as a parameter denoted by $\bar{\rho}$. Surfaces of section in the P_ϕ,ϕ plane can now be constructed numerically by strobing the dynamics at times $t_n = 2\pi n/\omega$ at various values of the polarization angle α. This is done for four values of α in Fig. 2 and, somewhat arbitrarily, $\omega = \bar{\rho} = F = 1.0$. In the CP limit the restricted problem is clearly integrable since the time dependence in the Hamiltonian vanishes. In particular for high angular momentum, circulating states [10] m is apparently a good quantum number in this approximation. Approximate conservation of m in the exact dynamics [12] confirms the applicability of the adiabatic approximation. As evidenced by Fig. 2, in an EP field a chaotic layer starts to develop for relatively small, negative values of P_ϕ. The asymmetry of the plots about $P_\phi = 0$ is a consequence of working in the rotating frame. In the nonrotating frame it has been

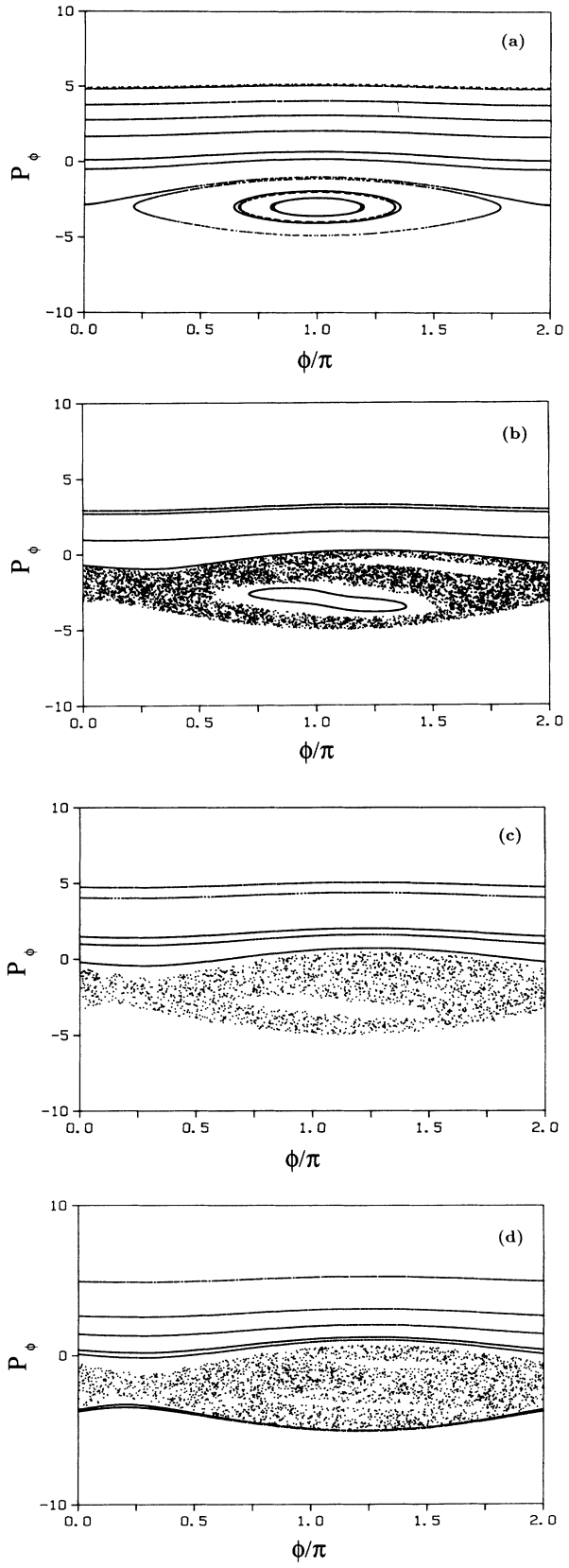

FIG. 2. Surfaces of section for the approximate Hamiltonian of Eq. (12) at various values of the polarization angle α; (a) $\alpha = 90°$, (b) $\alpha = 65°$, (c) $\alpha = 45°$, and (d) $\alpha = 25°$. The plots are all in a uniformly rotating frame as defined in the text.

verified numerically that the surfaces of section are essentially equivalent but, as expected, are symmetric about $P_\phi=0$. The degree of chaos grows with the ellipticity of the field, although the high angular momentum states continue to conserve m approximately. The presence of chaos is expected to lead to enhanced ionization rates since diffusion in the principle action n is now possible, through breakdown of the adiabatic approximation and strong coupling to the ρ mode. This corresponds to high-lying, bound, eccentric orbits for which the dynamics is driven into chaos when the electron's angular momentum in the nonrotating frame is relatively small along the orbit. A similar mechanism was proposed for the CP case [6] except that now the EP field accentuates ionization. Further study of this system is needed experimentally, numerically, and analytically to establish how accurately threshold field strengths are predicted as a function of polarization.

There has been recent interest in studying the microwave ionization of the hydrogen atom by two LP fields of different frequencies [13,14], because the second field can promote diffusion between resonance zones otherwise separated by Kolmogorov-Arnold-Moser surfaces [15]; it would be of considerable interest to investigate both theoretically and experimentally the effect of bichromatic fields of various polarizations on ionization rates. This would yield further insight into the mechanism of ionization in EP fields in terms of the overlap of classical resonance zones.

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