Stochastic instability of an oscillator and the ionization of highly-excited atoms under the action of electromagnetic radiation

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A new mechanism is proposed for the ionization of atoms highly excited by electromagnetic radiation.

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Processes of nonlinear photoionization of atoms are currently being studied intensely both experimentally and theoretically.[1] This note investigates the perturbation of the spectrum and proposes a new mechanism for the cascade ionization of highly-excited atoms (specificially hydrogen) under the action of a linearly-polarized mono-

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chromatic radiation with a frequency of $\omega \ll E_n^{(0)}$, where $E_n^{(0)}$ is the binding energy of the electron.[1] This mechanism is based on the classical effect of stochastic instability of a nonlinear oscillator.[2]

The condition $n \gg 1$ (n is the principal quantum number that corresponds to the canonical momentum of the classical Keplerian motion of the electron) permits applying a quasi-classical approximation. The Hamiltonian of the classical motion of the electron in the atom under the action of an electric field $\epsilon \cos\omega t$ has the form:

$$H = -\frac{1}{2n^2} + \epsilon \cos \omega t \left(1 - \frac{M_z^2}{M^2}\right)^{1/2} \sum_{k=1}^{\infty} (x_k \sin\psi \cos k\lambda + y_k \cos\psi \sin k\lambda) \tag{1}$$

Here, λ is the phase conjugated by n (in the unperturbed case, $\epsilon = 0$ $\lambda = \Omega t + \lambda_0$, where $\Omega = n^{-3}$ is the frequency of Keplerian motion of the electron), ψ and ϕ are the Euler angles,[3] and M and M_z are their conjugate momenta (the moment of the electron and its projection on ϵ). Because phase ϕ is cyclical, $M_z = \text{const}$. Further, x_k and y_k are the Fourier components of the dipole moment of the electron in a hydrogen atom.[4]

If the field intensity of wave ϵ is sufficiently small (see the criteria below), it is possible to distinguish between nonresonance and resonance. In the first case $|\omega - k\Omega| \sim \Omega$, the motion of the electron is determined in accordance with the conventional perturbation theory, which makes it possible to described the small "fluctuations" of the electron near the initial values of n and M. In the resonance case $|\omega - k\Omega| \ll \Omega$ and the Fourier series (Eq. 1) contains only the resonance term. Then, a simple canonical transformation eliminates the explicit temporal dependence of the Hamiltonian in Eq. (1). The relatively small size of the resulting displacement $\Delta n \ll n$ makes it possible to expand the first term in Eq. (1) close to the resonance value $n = n_k$ with an accuracy up to the terms $\sim (n - n_k)^3$, where $n_k = (k/\omega)^{1/3}$. The pairs of variables λ, n and ψ, M are the fast and slow subsystems, respectively, which makes it possible to bring Eq. (1) to the following form

$$H \approx -\frac{3}{2}\left(\frac{\omega}{k}\right)^{4/3}(n - n_k)^2 + \frac{\epsilon}{2}\left(1 - \frac{M_z^2}{M^2}\right)^{1/2} \times (x_{kr}^2 \sin^2\psi + y_{kr}^2 \cos^2\psi)^{1/2} \sin(k\lambda + \Phi), \tag{2}$$

where

$$x_{kr} \equiv x_k(n_k), \quad y_{kr} \equiv y_k(n)_k, \quad \text{tg}\Phi = (x_{kr}/y_{kr})\,\text{tg}\psi, \quad \psi = \text{const}, \; M = \text{const}.$$

The above Hamiltonian (Eq. 2)) is formally equivalent to the Hamiltonian of a mathematical pendulum of finite amplitude, while the equations of motion are integrated in elliptic functions.[5] The characteristic "beat" frequency of both phase (λ)-"captured" and fly-by particles is

$$\nu = \frac{\sqrt{3}}{2} k^{1/3} \epsilon^{1/2} \omega^{2/3} \left(1 - \frac{M_z^2}{M^2}\right)^{1/4} (x_{kr}^2 \sin^2\psi + y_{kr}^2 \cos^2\psi)^{1/4}. \tag{3}$$

For $k = 1$, Eq. (3) yields $\nu_{\max} = (3\epsilon)^{1/2}/2n$. The range of beats of n equals

$$\Delta n = \frac{4}{\sqrt{3}}\left(\frac{k}{\omega}\right)^{2/3} \epsilon^{1/2}\left(1 - \frac{M_z^2}{M^2}\right)^{1/4} (x_{kr}^2 \sin^2\psi + y_{kr}^2 \cos^2\psi)^{1/4}. \quad (4)$$

Substituting the resultant functions $n(t)$ and $\lambda(t)$ into Eq. (2), we may determine $M(t)$, $\psi(t)$ and $\phi(t)$ Quantization of the arbitrarily-periodic motion investigated[6] permits determination of the quasi-energetic state (QES) in both the non-resonant and resonant case. A separate article by the authors will be devoted to these findings.

Approximation of the isolated resonance is valid when $\nu \ll \Omega$,[2] i.e., $\epsilon \ll n^{-4}$ (let us recall that $\omega \ll n^{-2}$). Rigorous examination[7] has shown that when $\nu \ll \Omega$, for most initial conditions Δn remains small right up to $t = \infty$. For the remaining initial conditions, the "exit" velocity of n (the Arnold diffusion) is exponentially small.[8] Thus, in the classical formulation when $\epsilon \ll n^{-4}$, ionization is practically nonexistent.[1]

An erroneous assertion was expressed in a recent work[10] concerning the "diffusion" ionization of a hightly-excited atom in the case $\epsilon \sim n^{-5} \ll n^{-4}$. Without dwelling on the specific errors, let us note that the estimated "diffusion time" (Eq. (9) in Ref. 10) is classical. However, application of the corresponding results[10] to celestial mechanics would lead to the conclusion of the rapid escape of, for example, Saturn from the solar system occasioned by Jupiter, which contradicts observations.

The stochastic instability of an atomic oscillator and, as a consequence, ionization, occur when ν/Ω is not very small. In this case, various resonances overlap, while the separation of electrons into resonant and non-resonant electrons loses its meaning.[2] Moreover, the electron drifts chaotically through the levels until it returns to the continuum. The stochasticity threshold for systems with a Hamiltonian close to Eq. (1) is presented in Ref. 11 and has the form $\nu \gtrsim \Omega/6$ which, in our case, is equivalent to $\epsilon \gtrsim \epsilon_0 = 1/27n^4$. When $n = 66$, we get $\epsilon_0 = 10$ V/cm, which corresponds with the experimental results. A rough estimate of the ionization time (drifting of the electron among the resonances) close to the instability threshold yields $\tau \sim 10^3 n^3$. For $n = 50$, we find $\tau \sim 3 \times 10^{-9}$ sec, which corresponds with the experiments.[13] The presence of maxima in the dependence of the ionization probability on ω[13] can be explained by the incomplete overlapping of the resonance close to the stochasticity threshold.

Thus, the proposed mechanism of stochastic ionization apparently plays a significant role and must be taken into account when formulating a sequential quantum theory. After completion of this work, Ref. 14 appeared; it contains results of numerical modeling, by the Monte Carlo method, of the ionization and perturbation of hydrogen atoms for the experimental conditions.[12] In Ref. 14 the classical trajectories of electrons were computer-calculated. The good agreement of results of "classical" modeling[14] with experiment[12] is an additional agrument in support of our theory. It is important to mention that an appreciable portion of the trajectories obtained in Ref. 14 manifest properties of stochasticity, which was not noted by the authors of Ref. 14.

[1]Here and elsewhere, atomic units are used.

[2]Note that for $\epsilon \ll n^{-4}$, the corresponding quantum-mechanical problem is unsolved, with the exception of the region $\omega \ll \Omega$ (where the probability of ionization is exponetially small).[9]

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