Classical Self-Ionization of Fast Atomic Ions in Magnetic Fields

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In the presence of a magnetic field a permanent exchange of energy between the center of mass and electronic degrees of freedom of the atomic ion is shown to take place. For regular center of mass and internal motion we observe a self-stabilization of the highly excited ion on a Larmor orbit. For large values of the initial center of mass velocity the energy transfer from the center of mass to the electronic degrees of freedom is strong enough to allow the atom to ionize. This dynamical self-ionization effect is studied in some detail. Although the study is based on classical mechanics, it is argued that the effect should be observable in laboratory experiments.

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The behavior and properties of atoms in strong magnetic fields became in the past twenty years a field of great activity. The paradigm of this development was the hydrogen atom whose spectrum and eigenfunctions are today known to a high accuracy for Rydberg states up to and even beyond the field-free ionization threshold. The strong interest in the hydrogen atom was, in particular, motivated by the fact that it is one of the simplest physical systems which exhibits a transition from regularity to chaos both classically and quantum mechanically and which allows a detailed comparison of theoretical and experimental data [1].

In the presence of an external magnetic field the collective, i.e., center of mass (CM), and internal degrees of freedom of, for example, a two-body system cannot be separated. The constants of motion associated with the CM motion of the system are the components of the so-called pseudomomentum [2]. For a neutral system these components commute and a complete elimination of the CM coordinates from the Hamiltonian is possible [2,3]. Nevertheless, the CM and internal motion remain intimately coupled and, therefore, any change in the internal motion has drastic consequences for the behavior of the CM. Effects due to the inherent two-body character of a neutral system in a magnetic field have been investigated very recently for the hydrogen atom. In particular, for the case of vanishing pseudomomentum it has been observed that the transition from regularity to chaos in the classical internal motion is accompanied by a transition from bounded quasiperiodic oscillations to an unbounded diffusional motion in the CM [4]. For large enough values of the pseudomomentum (or equivalently an external static electric field) an outer potential well is formed supporting strongly delocalized bound states which have been studied in detail [5–7]. By performing a gauge independent pseudoseparation of the center of mass motion it was very recently shown in Ref. [7] that the potential leading to the formation of the above mentioned outer potential well is gauge independent.

In the present Letter we focus on charged atomic systems and particularly on the dynamics of the interaction of the CM and internal motion for the He^+ ion in an external homogeneous magnetic field. For a charged system the components of the pseudomomentum do not commute and, therefore, cannot be used for a complete elimination of the CM coordinates from the Hamiltonian. Nevertheless, the pseudomomentum is still useful for defining a unitarian which transforms the Hamiltonian to a particular simple and physically appealing form [8]. The transformed Hamiltonian for the He^+ ion takes on the following appearance:

$$\mathcal{H} = \mathcal{H}_1 + \mathcal{H}_2 + \mathcal{H}_3, \tag{1}$$

where

$$\mathcal{H}_1 = \frac{1}{2M}\left(\mathbf{P} - \frac{Q}{2}\,\mathbf{B}\times\mathbf{R}\right)^2, \tag{1a}$$

$$\mathcal{H}_2 = \alpha\,\frac{e}{M}\left[\mathbf{B}\times\left(\mathbf{P} - \frac{Q}{2}\,\mathbf{B}\times\mathbf{R}\right)\right]\mathbf{r}, \tag{1b}$$

$$\mathcal{H}_3 = \frac{1}{2m}\left(\mathbf{p} - \frac{e}{2}\,\mathbf{B}\times\mathbf{r} + \frac{Q}{2}\,\frac{m^2}{M^2}\,\mathbf{B}\times\mathbf{r}\right)^2 + \frac{1}{2M_0}\left\{\mathbf{p} + \left[\frac{e}{2} - \frac{Q}{2M}\,\frac{m}{M}\,(M + M_0)\right]\mathbf{B}\times\mathbf{r}\right\}^2 - \frac{2e^2}{r}, \tag{1c}$$

where m, M_0, and M are the electron, nuclear, and total mass, respectively. $\alpha = (M_0 + 2m)/M$ and Q is the net charge of the ion. $\mathbf{B}$ is the magnetic field vector which is assumed to point along the z axis. $(\mathbf{R},\mathbf{P})$ and $(\mathbf{r},\mathbf{p})$ are the canonical pairs for the CM and internal motion, respectively. The Hamiltonian $\mathcal{H}$ involves five degrees of freedom since the center of mass motion parallel to the magnetic field is a free translational motion, i.e., can be separated completely.

The Hamiltonians $\mathcal{H}_1$ and $\mathcal{H}_3$ contain exclusively the CM and internal degrees of freedom, respectively. $\mathcal{H}_1$ describes the free motion of a pseudoparticle, which corresponds to the ion as an entity, with charge Q and mass M in a magnetic field. $\mathcal{H}_2$ contains the coupling

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between the CM and internal motion of the ion and represents a Stark term with a rapidly oscillating electric field determined by the intrinsic dynamics. It is the interaction Hamiltonian $\mathcal{H}_2$ which is responsible for the effects and phenomena which will be investigated and discussed throughout this Letter.

In the following we present the results of a detailed study of the classical dynamics for the He^+ ion by solving the corresponding coupled Newtonian equations of motion belonging to the Hamiltonian $\mathcal{H}$. This is a nontrivial task since the long integration time necessary to observe the different time scales occurring in the corresponding dynamics (see below) require a fast integration algorithm with a very high accuracy and large step size. To achieve these goals we first performed the so-called Kustaanheimo-Stiefel transformation [9] in order to get rid of the singularity due to the Coulomb potential term. For the regularization of the CM degrees of freedom we took further advantage of the conservation of the pseudomomentum. For the integration of the resulting smoothened equations of motion we used a Bulirsch-Stoer integration algorithm which proved to be very fast and of an extremely high precision. Details of the overall procedure will be given elsewhere. We would like to stress that without such a procedure this study would not have been possible.

There are several characteristic regimes of the motion of the He^+ ion in a magnetic field, of which we describe here the three most interesting. Two frequently used quantities throughout this Letter are the CM kinetic energy $E_{\mathrm{CM}} = (M/2)\dot{\mathbf{R}}^2$ and the internal energy $E_{\mathrm{int}} = (\mu/2)\dot{\mathbf{r}}^2 - 2e^2/r$, where $\mu = mM_0/M$. With the help of the equations of motion it is possible to derive the following equation for the time dependence of the kinetic energy of the CM:

$$\frac{d}{dt} E_{\mathrm{CM}} = e\alpha(\mathbf{B} \times \dot{\mathbf{R}})\dot{\mathbf{r}}, \tag{2}$$

Extremal values of the CM energy therefore occur if the perpendicular components ($\perp \mathbf{B}$) of the CM and internal velocities are parallel, i.e., $\dot{\mathbf{r}}_\perp \parallel \dot{\mathbf{R}}$, whereas a strong flow of energy occurs for the orthogonal configuration, i.e., $\dot{\mathbf{r}}_\perp \perp \dot{\mathbf{R}}$.

Let us first investigate the regime for which the complete phase space is regular. The internal energy and field strength are chosen so that the Coulomb potential dominates strongly over the magnetic interaction terms. In particular, we concentrate on the subset of initial conditions with vanishing CM velocity, i.e., $V_{\mathrm{CM}} = |\dot{\mathbf{R}}| = 0$. In the complete absence of a magnetic field the ion would simply be at rest. In the presence of the magnetic field, however, the coupling term $\mathcal{H}_2$ induces an oscillating flow of energy between the CM and internal degrees of freedom [see Eq. (2)]. We observe now four, by orders of magnitude different, time scales for the CM motion. The shortest time scale is that of a single oscillation of the CM energy and motion which corresponds to one slightly perturbed Kepler cycle in the internal motion. The second time scale arises due to the electronic Zeeman term which causes a rotation of the perturbed Kepler ellipses. The third time scale occurs because of the quasiperiodic evolution of the orbital parameters of the ellipses of the internal motion [10] and due to the action of the coupling Hamiltonian $\mathcal{H}_2$. We find a slow oscillatory modulation of the CM and internal motions (energy) on top of the above mentioned faster motions. Finally, on the fourth and longest time scale the CM performs a circular motion which can be shown to be the motion of a free pseudoparticle with charge Q and mass M in a magnetic field with Larmor frequency $\omega_L = QB/M$. In spite of the fact that the initial CM velocity of the ion was equal to zero we encounter on the longest time scale the effect of self-stabilization of the ion on a Landau orbit. The natural question now arises for the Larmor radius of this orbit. Since we refer to a pseudoparticle picture (which is reasonable if the Coulomb potential dominates the magnetic interaction), the pseudomomentum gives us the coordinates of the center of the Landau orbit of the CM. With the help of the equations of motion we arrive at the following expression for the Larmor radius R_L:

$$R_L = \left| \frac{M}{QB^2} \mathbf{B} \times \dot{\mathbf{R}}(0) - \frac{e}{Q} \alpha \mathbf{r}_\perp(0) \right|, \tag{3}$$

where we have assumed without loss of generality that $\mathbf{R}(0) = \mathbf{0}$. For the above discussed situation this means that the Larmor radius of the CM motion is completely determined by the initial distance between the electron and the nucleus in the plane perpendicular to the magnetic field. All amplitudes of the oscillations on the above mentioned shorter time scales are small compared to this Larmor radius. We remark that the above discussed effect of the classical self-stabilization of the ion on a Landau orbit is a generic phenomenon for regular phase space, i.e., it occurs for any regular initial condition.

Next, we turn to the regime of motion where the *internal* phase space would be completely chaotic if the nuclear mass were infinite. The initial internal energy and/or the field strength are chosen to fulfill this condition. The internal CM velocity is chosen such that the total energy is close to the threshold $E = 0$. As a characteristic phenomenon of the resulting classical dynamics we observe intermittent behavior [6] of the CM as well as internal motion. After an initial phase of chaotic motion (with a nonvanishing local Lyapunov exponent [11,12]) and an oscillating flow of energy from and to the CM (internal) motion, a sudden strong energy transfer form the CM to the internal degrees of freedom takes place. As a consequence, the available phase space for the internal motion is enlarged and a quasiregular phase of large amplitude internal motion follows, during which the magnetic interaction dominates the Coulomb potential. The quasiregular phase ends with a sudden energy transfer from the internal degrees of freedom back to the CM motion and

consequently another chaotic phase of motion takes place. This phenomenon occurs repeatedly.

The most interesting dynamics of the ion occurs if we increase the values of the initial CM kinetic energy. The initial internal energy again corresponds to a bound Rydberg state of the ion, whereas the total energy is now considerably above zero. After a transient time of bound oscillations in the internal motion (energy), a strong flow of energy from the CM to the internal motion takes place. The *internal* energy is hereby increased above the threshold for ionization, $E_{\text{int}} = 0$, and the ion eventually ionizes, i.e., the electron escapes in the direction parallel to the magnetic field. Note that the motion of the electron is confined in the direction perpendicular to the magnetic field.

Figure 1 provides a prototype example for such an ionizing trajectory. Figures 1(a) and 1(b) illustrate the time dependencies of the CM energy and the z component of the internal relative coordinate, respectively. After the above mentioned initial phase of oscillations there occurs at approximately $T = 1.7 \times 10^{-10}$ s a sudden loss of CM kinetic energy simultaneously accompanied by an increase in the internal energy which causes the electron to move away from the nucleus in the positive z direction. The transferred energy, which is in our case of Fig. 1 approximately 0.2 eV, corresponds only to a small fraction of the total initial CM energy which is for our example about 333.8 eV. This energy transfer is only possible due to the presence of the coupling term $\mathcal{H}_2$ in the Hamiltonian (1) which involves both the internal and CM degrees of freedom. The ionization time for an individual trajectory depends, apart from its intrinsic dynamics, on the field strength and in particular on the CM kinetic energy of the ion.

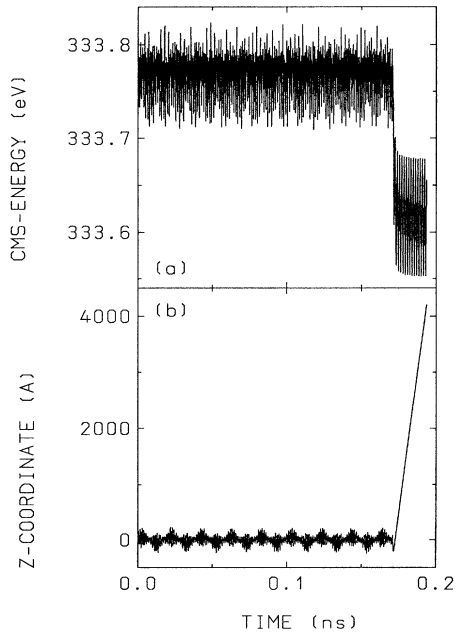

FIG. 1. (a) The CM energy as function of time. (b) The z component of the internal relative coordinate as a function of time. The total and initial internal energy of the ionizing trajectory are $E = 333.68$ eV and $E_{\text{int}} = -8.16 \times 10^{-2}$ eV, respectively. The field strength is $B = 23.5$ T.

In order to obtain a statistical measure for the ionization process we calculated for an ensemble of trajectories the fraction of ionized orbits as a function of time. The *initial internal energy* was chosen to correspond to a completely chaotic phase space of the internal motion of the He^+ ion if the nuclear mass were infinite, i.e., the Coulomb interaction and magnetic energies are of equal order of magnitude. The initial conditions for the internal motion have therefore been selected randomly on the energy shell. In Fig. 2 we have illustrated the fraction of ionized orbits as function of time up to $T = 2.4 \times 10^{-7}$ s, for a series of different energies and for a fixed laboratory field strength of $B = 23.5$ T. For an initial CM energy of $E_{\text{CM}} = 1.45$ eV, which corresponds to an initial CM velocity of $V_{\text{CM}} = 8.4 \times 10^3$ m/s, about 70% of the trajectories are ionized within a time of $T = 2.4 \times 10^{-8}$ s, which is the tenth part of the integration time. In contrast to this we have for $E_{\text{CM}} = 0.27$ eV only about 30% of the ionized orbits within the total integration time of $T = 2.4 \times 10^{-7}$ s. The ionization process depends very sensitively on the initial CM kinetic energy of the ion. We remark that the ionization fractions plotted are independent of the chosen gauge since they arise from the classical trajectories which are solutions of the Newtonian equations of motion which, themselves, are gauge independent.

The above discussed self-ionization effect occurs also for initial internal energies and field strengths which belong to the regular regime. However, in this case we need much higher initial CM velocities in order to obtain a

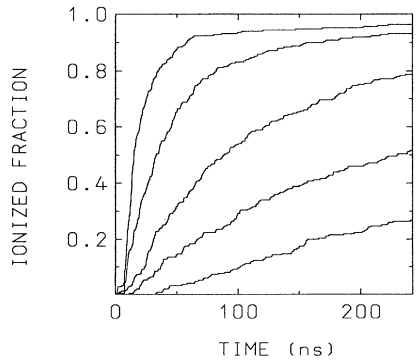

FIG. 2. The ionized fraction for an ensemble of 250 trajectories as a function of time. From top to bottom, the CM energies belonging to the ionization curves are $E_{\text{CM}} = 1.45$, 0.63, 0.47, 0.34, and 0.27 eV, respectively. The initial internal energy is always $E_{\text{int}} = -9.25 \times 10^{-3}$ eV. The field strength is $B = 23.5$ T.

substantial ionization rate. For example, for an initial CM kinetic energy, $E_{CM} = 27.3$ eV ($V_{CM} = 3.6 \times 10^4$ m/s), and an initial internal energy, $E_{int} = -8.16 \times 10^{-2}$ eV, about 13% of the trajectories are ionized within $T = 2.4 \times 10^{-8}$ s, whereas for the same internal energy and $E_{CM} = 109$ eV ($V_{CM} = 7.25 \times 10^4$ m/s) about 80% of the orbits are ionized within the same time interval. The CM kinetic energy necessary to observe ionization increases rapidly with decreasing internal energy and also with decreasing strength of the external magnetic field. We remark that the above values of the CM energies and velocities are well within the range of energies and velocities for which a nonrelativistic approach to the dynamics of the ion is valid.

All the considered values for the initial internal energy correspond to highly excited Rydberg states of the He^+ ion in a strong magnetic field. For sufficiently high excited electronic states the perturbation due to the coupling Hamiltonian $\mathcal{H}_2$ becomes lager than the spacing of adjacent levels of the internal Hamiltonian $\mathcal{H}_3$ [8]. As a consequence, strong mixing of the electronic and CM wave functions occurs. This quantum regime of mixing includes the above discussed classical regime for which we observe the process of self-ionization of the ion. Since we are dealing with highly excited states for which the action is much larger than the elementary quantum of action, we expect the self-ionization mechanism of the ion to survive quantization. This should have implications on different areas of physics like plasma or astrophysics for which the stability of highly excited charged atoms in strong magnetic fields (at finite temperatures) is a relevant question. In particular, the self-ionization phenomenon lends itself to laboratory experiments. In order to observe the energy transfer from the CM to the electronic states of the atomic ion under consideration we suggest the following experiment: A fast beam of atomic ions has to be injected in a homogeneous magnetic field and subsequently the ions are excited by photons whose appropriately chosen frequency is below the threshold energy for ionization. Nevertheless, ionization, i.e., electron emission in the direction parallel to the magnetic field, should be observable by transfer of energy from the CM to the electronic motion. We remark that the typical electromagnetic decay time of the Rydberg states of the ion is of the same order of magnitude as our typical ionization times. They are therefore competing processes. For stronger fields and/or larger CM velocities the self-ionization process dominates. Finally we mention that very recently the effect of stabilization for neutral atoms in crossed static fields has been observed [13] for a certain range of magnetic field strengths. For atoms in a microwave field, stabilization as well as enhanced ionization can occur [14]. If at all, the present case of an ion in a magnetic field resembles the latter case because the coupling of the center of mass and internal electronic degrees of freedom for an ion in a magnetic field implies a rapidly oscillating intrinsic electric field.

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[1] H. Friedrich and D. Wintgen, Phys. Rep. **183**, 37 (1989).
[2] J. E. Avron, I. W. Herbst, and B. Simon, Ann. Phys. (N.Y.) **114**, 431 (1978).
[3] B. R. Johnson, J. O. Hirschfelder, and K. H. Yang, Rev. Mod. Phys. **55**, 109 (1983).
[4] P. Schmelcher and L. S. Cederbaum, Phys. Lett. A **164**, 305 (1992); Z. Phys. D **24**, 311 (1992).
[5] D. Baye, N. Clerbaux, and M. Vincke, Phys. Lett. A **166**, 135 (1992).
[6] P. Schmelcher and L. S. Cederbaum, Chem. Phys. Lett. **208**, 548 (1993); Phys. Rev. A **47**, 2634 (1993).
[7] O. Dippel, P. Schmelcher, and L. S. Cederbaum, Phys. Rev. A **49**, 4415 (1994).
[8] P. Schmelcher and L. S. Cederbaum, Phys. Rev. A **43**, 287 (1991).
[9] E. L. Stiefel and G. Scheifele, *Linear and Regular Celestial Mechanics* (Springer-Verlag, Berlin, 1971).
[10] J. B. Delos, S. K. Knudson, and D. W. Noid, Phys. Rev. A **28**, 7 (1983).
[11] H. Fujisaka, Prog. Theor. Phys. **70**, 1264 (1983).
[12] P. Grassberger and I. Procaccia, Physica (Amsterdam) **13D**, 34 (1984).
[13] G. Raithel and H. Walther, Phys. Rev. A **49**, 1646 (1994).
[14] G. Casati, I. Guarneri, and D. L. Shepelyansky, IEEE J. Quantum Electron. **24**, 1420 (1988).