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Mesoscopic motion of atomic ions in magnetic fields

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We introduce a semiclassical model for highly excited atomic ions moving in a magnetic field, which allows us to describe the mixing of the Landau orbitals of the center of mass in terms of the electronic excitation and magnetic field. The extent of quantum energy flow in the ion is investigated and a crossover from localization to delocalization with increasing center of mass energy is detected. Our model of the moving ion in a magnetic field turns out to be closely connected to models for transport in disordered finite-size wires. [S1050-2947(98)50611-7]

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Interacting particle systems in strong magnetic fields show a rich variety of complex phenomena. The source of this complexity is competition between the magnetic and Coulomb interactions, which are of inherently different character. With changing strength of the external field the corresponding systems undergo a metamorphosis involving qualitatively different states. In atomic physics attention focused for more than a decade on the hydrogen atom in a magnetic field (see Ref. [1] and references therein) on which detailed experimental and theoretical investigations yielded many excellent insights into semiclassical and quantum aspects of nonintegrable systems and significantly enhanced our understanding of the new features arising due to the presence of the external field. With an increasing degree of excitation and/or increasing field strength the electronic motion of the classical atom shows a transition from regular to irregular, i.e., chaotic behavior and intermittency. More recently it has become evident that the nonseparability of the collective, i.e., center of mass (c.m.) and electronic motion of atoms in the presence of a magnetic field, leads to a variety of two-body phenomena. The corresponding coupling of the c.m. and electronic motion is fundamentally different for neutral and charged systems. Examples of two-body effects in neutral systems are the classical chaotic diffusion of the c.m. [2] or the existence of weakly bound giant dipole states [3]. For atomic ions the interaction of the c.m. and electronic degrees of freedom is more intricate and manifests itself in a continuous, classical flow of energy from the collective to the internal motion and vice versa. Detailed studies of the classical dynamics of rapidly moving highly excited He^+ ions in a magnetic field showed that this energy exchange leads to the self-ionization process [4] of the ion. Very little, however, is known about the quantum properties and behavior of moving highly excited atomic ions.

Using Landau orbitals for the c.m. motion in zeroth-order and fixed nucleus zero-field wave functions for the electronic motion to estimate their coupling matrix elements, it was demonstrated [5] that there exist a number of different physical situations for which the interaction between the collective and electronic motion becomes strong. The latter induces a strong mixing of the c.m. and electronic motion and is a potential source of interesting quantum properties of the ion, in particular when the dynamics of the corresponding classical ion is chaotic. A detailed investigation of the coupled c.m. and electronic motion of the highly excited quantum-mechanical ion in this regime, which is the subject of interest in the present paper, is, however, a highly nontrivial task: we are dealing with five nonseparable and strongly mixing degrees of freedom in a regime of very high level density, which depends on a number of parameters (field strength, total energy, etc.). The *ab initio* description of the quantum dynamics in the above regime goes even beyond modern computational possibilities and we thus seek a model approach that captures the essential physics of the problem. Here we propose and analyze a semiclassical model of the excited ion, and explore the consequences of coupling between its c.m. and electronic degrees of freedom.

Since we deal with the interaction of the c.m. and electronic motion in atomic ions we first have to introduce collective (c.m.) and relative variables in the Hamiltonian describing the atom. The total pseudomomentum $\boldsymbol{K}$ [6] is a conserved quantity associated with the c.m. motion, which, in spite of the fact that its components perpendicular to the magnetic field are not independent, i.e., do not commute, can be used to transform the Hamiltonian to a particularly simple and physically appealing form [5], which for the He^+ ion reads $\mathcal{H}=\mathcal{H}_1+\mathcal{H}_2+\mathcal{H}_3$ where

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

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

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

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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. $\boldsymbol{B}$ is the magnetic field vector, which is assumed to point along the z axis. $(\boldsymbol{R},\boldsymbol{P})$ and $(\boldsymbol{r},\boldsymbol{p})$ are the canonical pairs for the c.m. and internal motion, respectively. $\mathcal{H}$ involves five degrees of freedom, since parallel to the magnetic field the c.m. undergoes free translational motion and can be separated completely.

$\mathcal{H}_1$ and $\mathcal{H}_3$ depend exclusively on the c.m. and electronic degrees of freedom, respectively. $\mathcal{H}_1$ describes the free motion of a c.m. pseudoparticle with charge Q and mass M. $\mathcal{H}_3$ describes the electronic motion in the presence of paramagnetic, diamagnetic, as well as Coulomb interactions, which, analogous to the hydrogen atom [1], exhibits an enormous complexity of classical and quantum properties with changing parameters, i.e., energy and/or field strength. $\mathcal{H}_2$ contains the coupling between the c.m. and electronic motion of the ion and represents a Stark term with a rapidly oscillating electric field $1/M\,(\boldsymbol{B}\times[\boldsymbol{P}-(Q/2)\boldsymbol{B}\times\boldsymbol{R}])$ determined by the dynamics of the ion. It is the interaction Hamiltonian $\mathcal{H}_2$ that is responsible for the interesting quantum effects that will be investigated in the present Rapid Communication.

From the above it is natural to consider the representation of the coupling Hamiltonian $\mathcal{H}_2$ in a basis that consists of products of eigenstates $\Phi_{\text{c.m.}}$ of $\mathcal{H}_1$ and Ψ of $\mathcal{H}_3$. Calculating the corresponding matrix elements we encounter some selection rules that are of immediate relevance to our model (see below). Since the total angular-momentum component parallel to the magnetic field $\mathcal{L}_z$ is a conserved quantity for $\mathcal{H}$, and since the corresponding c.m. angular-momentum $L_{\text{c.m.}_z}$ and electronic angular-momentum L_z are conserved quantities for $\mathcal{H}_1$ and $\mathcal{H}_3$, respectively, the matrix elements of $\mathcal{H}_2$ involve only c.m. and electronic states with magnetic quantum numbers that are correspondingly different. In addition, we have the relation $\langle\Phi'_{\text{c.m.}}|[\boldsymbol{P}-(Q/2)\boldsymbol{B}\times\boldsymbol{R}]|\Phi_{\text{c.m.}}\rangle = iM(E'-E)\langle\Phi'_{\text{c.m.}}|\boldsymbol{R}|\Phi_{\text{c.m.}}\rangle$, which, together with the dipole selection rules for electronic transitions, allows only changes of the c.m. (μ) and electronic (m) magnetic quantum numbers by 1 and requires a change of energy for the c.m. motion. According to Ref. [5] the matrix elements of $\mathcal{H}_2$ involve a factor $\sqrt{N}$ for $N\gg|\mu|$. (N is the Landau principal quantum number of the c.m. motion), which yields the scaling of the coupling matrix elements of $\mathcal{H}_2$ with respect to varying c.m. energy.

Our model for the moving He^+ ion in a magnetic field is built up from three key constituents associated with the Hamiltonians $\mathcal{H}_1$, $\mathcal{H}_2$, and $\mathcal{H}_3$. The equidistant and infinitely degenerate spectrum of $\mathcal{H}_1$ is completely characterized by the c.m. quantum numbers N and μ. $\mathcal{H}_1$ represents the integrable part of the system that is coupled via $\mathcal{H}_2$ to the chaotic part represented by $\mathcal{H}_3$. The classical dynamics of $\mathcal{H}_3$ [7] depends on the scaled energy and angular momentum, which are given by $\hat{L}_z=L_z(\frac{1}{4}B)^{1/3}$ and $\hat{E}=E(2B)^{-2/3}$, respectively. In order to locate the regime of chaotic electronic motion we have made Poincaré surfaces of section of the classical dynamics of $\mathcal{H}_3$ $(\hat{E},\hat{L}_z)$ for a dense grid of values of the scaled energy and angular momentum. Starting with a completely chaotic phase space for $\hat{L}_z=0$ we find that with increasing values of the angular momentum the fraction of chaotic phase-space volume decreases rapidly. In contrast, the regime of negative $\hat{L}_z$ that yields predominantly chaotic phase space is much larger. If we take a typical scaled energy of $\hat{E}=-0.1$, for example, and require more than 90% of phase space to be occupied by chaotic trajectories, we obtain the regime $\hat{L}_z=[-2.71,0.136]$, which corresponds to $L_z=[-200,+10]$ for $B=10^{-5}$ (we use atomic units throughout, i.e., the field strength $B=1$ a.u. corresponds to 2.35×10^5 T). The mixing of electronic eigenfunctions belonging to negative values of the angular momentum L_z therefore represents an important ''open channel'' with respect to the coupling of the chaotic electronic motion to the c.m. motion. In this region of L_z the c.m. motion of the classical ion is strongly affected by coupling to the chaotic electron. Indeed, studies of the classical dynamics of the ion close to the ionization threshold demonstrated [4] that large negative values of the angular momentum are an inherent feature of intermittent dynamics as well as a prerequisite for the self-ionization process, whereby energy transfer from the c.m. to the electron results in ionization. We therefore turn our attention to this subspace and investigate the quantum-mechanical properties of this channel.

The spectrum belonging to the chaotic Hamiltonian $\mathcal{H}_3$ will be represented by a random matrix ensemble, which is the appropriate semiclassical description [9,10]. Since the Hamiltonian $\mathcal{H}_3$ possesses a generalized time-reversal invariance [8], which consists of a rotation by π around the x axis and a subsequent conventional time-reversal operation, the proper ensemble is the Gaussian orthogonal ensemble (GOE). While the GOE provides the fluctuations of the chaotic levels, we still need to specify the mean level density (MLD) as a function of the energy, field strength, and in particular the angular momentum L_z. Our approach to the MLD is via the semiclassical Thomas-Fermi formula. Performing the appropriate scale transformations, fixing $\hat{L}_z$, transforming to cylindrical coordinates, and subsequently performing the integrations over ϕ,p_z,p_ρ,z we arrive at the following result for the semiclassical MLD:

$$\rho_{L_z}(E,B)=(2B^{-2})^{2/3}2\int_0^\infty d\rho\sqrt{\mathcal{A}}\Theta(\mathcal{A}), \tag{2}$$

with $\mathcal{A}=[1/2\rho^2(\rho^2/2+\hat{L}_z)^2-\hat{E}]^{-2}-\rho^2$. The remaining integration over ρ has to be performed numerically. Starting with $L_z=0$ a general feature of the MLD is its rapid decrease with increasing L_z, whereas for negative values of L_z its decay is much weaker. Not only the fraction of chaotic phase space but also the absolute phase-space volume persist down to large negative values of the angular momentum, to $L_z\approx-250$, and therefore $\rho_{L_z}(E,B)$ represents in this regime the density of irregular states. Having specified our GOE, whose MLD at the center of the band is given by Eq. (2) providing the levels of $\mathcal{H}_3$, we turn to the calculation of the coupling matrix elements introduced by $\mathcal{H}_2$. The size of the matrix elements of $\mathcal{H}_2$ can be determined from a semiclassical relation between off-diagonal matrix elements of an operator and the Fourier transform of its classical autocorrelation function [11]. The variance $\sigma^2_{\mathcal{H}_2}$ of the matrix elements of $\mathcal{H}_2$ depends on the energies of the states they couple and

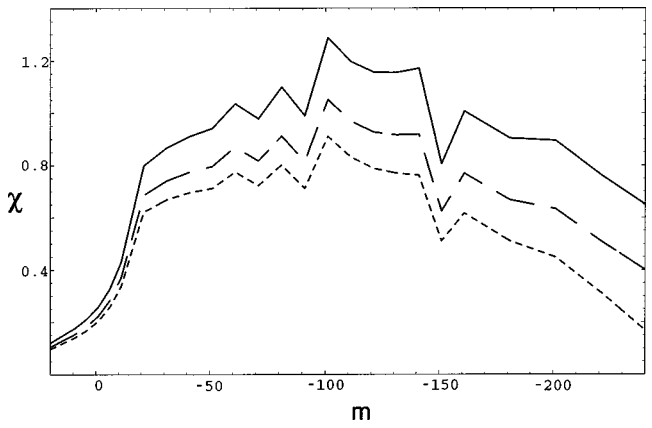

FIG. 1. The model parameter $\chi \equiv \rho_{L_z} |\sigma_{\mathcal{H}_2}|$, where ρ_{L_z} and $|\sigma_{\mathcal{H}_2}|$ are calculated using Eqs. (2) and (3), respectively, as a function of the angular-momentum quantum number m. At $m=0$, $N=250$ (short dashes), 300 (long dashes), and 400.

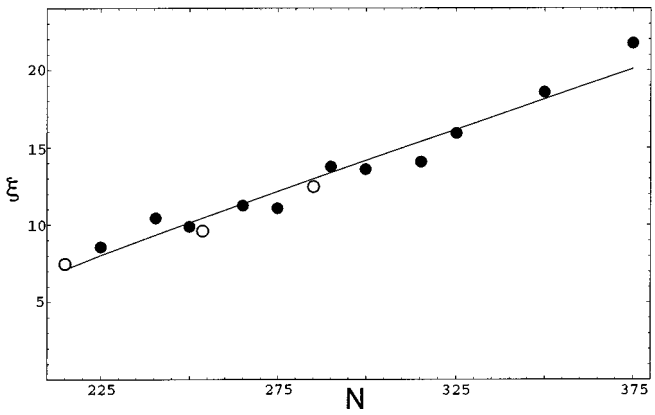

FIG. 2. Localization length computed for the ion at different values of the Landau level N at $m=0$, from which χ is computed with Eqs. (2) and (3). Filled circles are results for the ion model and open circles for the simpler version as described in the text. The line is a fit through the data.

is very small when the energy difference is greater than the level spacing of $\mathcal{H}_1$ [11]. For states close in energy, $\sigma^2_{\mathcal{H}_2}$ appears as

$$\sigma^2_{\mathcal{H}_2} \approx (4\pi\rho_{L_z})^{-1} \int_{-\infty}^{\infty} \langle \mathcal{H}_2(t)\mathcal{H}_2(0)\rangle e^{-i\omega t} dt, \qquad (3)$$

where selection rules determine the numerical coefficient of Eq. (3) [10]. Efficient evaluation of the above equation through classical trajectory calculations is by no means trivial and is done by introducing a suitable ensemble average via the periodogram approximation [12]. Figure 1 shows $\chi \equiv \rho_{L_z} |\sigma_{\mathcal{H}_2}|$ as a function of the angular momentum, calculated using Eqs. (1)–(3). As examples we plot $N=250$, 300, and 400 (at $m=0$) and $\mu<0$. Due to the selection rules N increases (decreases) by 1 as m decreases (increases) by 1. As seen in the figure, the largest values of χ lie mainly in the interval $m \approx [-20,-200]$.

Based on the structure of $\mathcal{H}$ the organization of our model is as follows. We have an array of sites, each of which corresponds to eigenstates of $\mathcal{H}_1$ and $\mathcal{H}_3$ labeled by particular values of N, m, and μ. We thus assign to each site the c.m. quantum numbers N,μ of a Landau orbital belonging to $\mathcal{H}_1$ and the levels of one member of the GOE representing the irregular levels of $\mathcal{H}_3$, which are labeled by m. The energy levels at each site are the $\mathcal{H}_3$ levels plus the level of $\mathcal{H}_1$. They are coupled by random matrix elements to levels of their neighboring sites, as imposed by the selection rules. The variance of the random elements $\sigma^2_{\mathcal{H}_2}$ is given by Eq. (3). The model thus has a one-dimensional structure, where sites labeled by N, m, and μ are comprised of levels taken to be from a GOE whose density is given by Eq. (2). The ion model so defined resembles the semiclassical pump model of Arnold diffusion [13]. The classical stochastic pump model describes Arnold diffusion in terms of the pumping of otherwise regular trajectories via weak, irregular motion within the Hamiltonian system [14]. Similarly, the classical c.m. motion of the ion is coupled to the chaotic motion of the electron due to the magnetic field. The semiclassical pump model of Arnold diffusion was found to be equivalent to models of single-particle transport in disordered wires, which predict localization [15–17]. The localization length of the semiclassical pump model was expressed in terms of corresponding classical parameters. Likewise, we should be able to predict any localization of the quantum ion in terms of the corresponding classical system.

The values of all the ion parameters (N,μ,E_3,L_z,B) are embodied in the model parameter $\chi \equiv \rho_{L_z} |\sigma_{\mathcal{H}_2}|$, examples of which are shown in Fig. 1. Though χ clearly fluctuates over the sites of the model, we could nevertheless estimate the localization length for this model in terms of an average, $\overline{\chi}$, over χ. As seen in Fig. 1, the range in m over which energy transfer can most appreciably occur is $m \approx [-20,-200]$, though the model also encompasses a wider range of m, where classically the ion is still predominately chaotic. In terms of the average $\overline{\chi}$, the localization length ξ_∞ of the model, assuming an infinite number of sites, is $\xi_\infty = 4\pi^2\overline{\chi}^2$ [13,16]. Because the ion model has a finite length of about 200 sites, the localization length can be estimated using finite-size scaling arguments for band random matrices, such as the ion model, since $\xi \approx \xi_\infty/(1+c\xi_\infty/L)$, $c \approx 1$ [18], where L is the length or the number of sites of the model. The N dependence of ξ arises from $\xi_\infty \sim N$, since $|\sigma_{\mathcal{H}_2}| \sim \sqrt{N}$ (see above). Solving for ρ_{L_z} and $|\sigma_{\mathcal{H}_2}|$ using Eqs. (2) and (3) we find, e.g., that $\xi \approx \xi_\infty \approx 30$ for $N=400$ ($m=0$). Thus, starting in a Landau level near $N \approx 400$, quantum flow is restricted to about 30 sites in m. This is in contrast to the classical ion, where, for a corresponding initial c.m. energy of $\approx 6\times10^{-7}$ a.u., there is no such restriction in the chaotic motion over L_z. Since χ varies as $\sqrt{N}$ the ion remains localized, in contrast to the classical ion, for N up to values near 4000. Taking the cyclotron frequency to be 1.4×10^{-9} a.u., this corresponds to a c.m. energy of about 6×10^{-6} a.u.

These arguments, while using well-known predictions for equivalent random matrix models, nevertheless depend on our being able to use an average of χ over the length of the model to estimate ξ. As a check, we have studied numerically the ion model to compare predictions of ξ using the actual χ, which varies as determined by the semiclassical results for the ion calculated with Eqs. (1)–(3), with results using $\overline{\chi}$, defined as the average χ over the length of the

model. Our numerical model ranges from $m=-230$ to 0. Each site consists of 12 levels of the GOE. Though this is a very small number, it is all we could include computationally while also incorporating the largest possible number of sites. We average over six realizations of each set of parameters and calculate the localization length of the eigenvectors. Results are shown in Fig. 2, where we see that both models, the ion and the simpler version with $\overline{\chi}$, give the same results for ξ, and are close to the line $\xi=0.72\xi_\infty$, where $\xi_\infty=4\pi^2\overline{\chi}^2$, and the factor 0.72 is purely an artifact of using only 12 levels per site, which is seen upon comparing with numerical results where more levels per site were used [13].

The occurrence of the crossover from localized to delocalized c.m. motion that we find for the ion at a c.m. energy of about 6×10^{-6} a.u. with the above parameters will of course vary with the strength of the field and the internal energy of the ion, as well as its mass, since we have considered only He^+ here. This crossover should be observable spectroscopically since the regimes of very weak and strong mixing show inherently different level spacings and absorption features. Finally, we mention that the above-investigated quantum mixing of collective and electronic motion for atomic ions is certainly of interest also for charged molecular systems in a magnetic field. Here the heavy vibrational and rotational degrees of freedom couple, for heteronuclear systems, to the collective motion of the molecular ion providing a potential source of new rotational and vibrational structures.

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