Giant dipole states of multielectron atoms in crossed electric and magnetic fields

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(Received 30 April 2001; published 16 November 2001)

Giant dipole states of singly and excited doubly multielectron atoms in crossed electric and magnetic fields are investigated. A gauge-independent approach to the pseudoseparation of the center of mass yields a generalized multielectron potential in crossed fields that serves as a basis for the study of the dipole states. For doubly excited systems, a class of highly symmetric decentered configurations is found and the properties of the corresponding resonances are determined. An outline on multiply excited systems is given.

DOI: 10.1103/PhysRevA.64.063412 PACS number(s): 32.60.+i

I. INTRODUCTION

External fields strongly influence the structural as well as dynamical properties of matter and, in particular, of atoms or molecules. Unexpected phenomena have been found for the most fundamental atomic system, i.e., the hydrogen atom exposed to magnetic and electric fields (see Refs. [1–4] and references therein). Beyond the specific phenomena induced by the external field, the hydrogen atom represents a paradigm of a complex and nonintegrable system, whose detailed understanding influenced a number of modern theoretical as well as experimental developments. Examples are the semiclassical theory of classically chaotic systems and the experimental technique called scaled energy spectroscopy.

More recently, it has become evident that the nonseparability of the center of mass (CM) and electronic motion for the hydrogen atom in a magnetic field leads to a variety of two-body phenomena [5] such as the chaotic diffusion of the CM [6,7]. Due to a gauge-independent approach to the separation of the CM and electronic motion of the atom, it was possible to prove [8] that there exists a generalized gauge-independent potential $\mathcal{V}$ for the electronic motion. In addition to the Coulomb potential, $\mathcal{V}$ contains linear and quadratic terms with respect to the electronic coordinates perpendicular to the magnetic field, plus a constant associated with the electric field being either of motional or external origin. If the electric field exceeds a certain threshold value, the combined action of these potential terms leads to the existence of an outer well, whose minimum is strongly shifted from the Coulomb singularity at the origin. This outer well accomodates typically a huge number of weakly bound states that possess, at laboratory field strengths, a very large electric dipole moment (so-called decentered states [9,10]) and which are inherently different from the ''traditional'' hydrogenic Rydberg states in a magnetic field located in the Coulomb well. For low-lying energetical excitations, the eigenstates in the outer well may be approximated by those of a charged anisotropic harmonic oscillator in a magnetic field. With an increasing degree of excitation, however, anharmonicity effects become relevant and ''exact'' numerical calculations are indispensable in order to gain insight into the properties of the system [8]. A detailed description of an experimentally feasible scheme allowing for the preparation of the hydrogenic giant dipole states was provided in Ref. [11]. It is based on the population of a Coulomb-Rydberg state by laser excitation, followed by a two-step switching of an external electric field, thereby capturing the electronic-nuclear relative motion in the outer well. In an application to matter-antimatter systems, it could be shown [12] that for the outer-well states of the positronium atom, the positron and electron are prevented from annihilation for time scales as long as several years, i.e., an isolated quasistable positronium atom exists in crossed fields.

In the present paper, we investigate giant dipole states of multielectron atoms in crossed electric and magnetic fields. To do this, we first derive in Sec. II, the generalized potential for neutral multielectron atoms by performing a gauge-independent pseudoseparation of their CM. Section III is devoted to the treatment of singly excited decentered atoms. Section IV contains an investigation of doubly excited systems that yields a class of resonances in crossed fields corresponding to highly correlated decentered configurations of the atoms.

II. THE GENERALIZED POTENTIAL FOR MULTIELECTRON ATOMS IN CROSSED FIELDS

Since the above-summarized results have been obtained in the framework of *one-electron atoms*, the question arises whether there exist decentered structures of *multielectron systems* and what their properties are. Since the gauge-independent approach to the separation of the CM is the key ingredient for the derivation of both the outer well as well as the giant dipole states, our first major step is to prove the existence of a generalized gauge-independent multielectron potential. To do so, we start with the atomic Hamiltonian in the presence of external magnetic and electric fields in the laboratory frame

$$\mathcal{H}=\frac{1}{2m}\sum_{i=1}^{N}(\mathbf{p}_i'-e\mathbf{A}_i')^2+\frac{1}{2M_0}(\mathbf{p}_0'+Ze\mathbf{A}_0')^2 - e\sum_{i=1}^{N}\mathbf{E}\cdot\mathbf{r}_i'+Ze\mathbf{E}\cdot\mathbf{r}_0'+V_c(|\mathbf{r}_i'-\mathbf{r}_j'|,|\mathbf{r}_i'-\mathbf{r}_0'|), \quad (1)$$

where the indices i,0 refer to the electrons and the nucleus, respectively. V_c contains all Coulomb interaction terms.

E, **A** are the electric field and vector potential, respectively. The (total) pseudomomentum $\hat{\mathbf{K}}$ is a constant of motion of the above Hamiltonian. It possesses the following gauge-independent appearance [13]

$$\hat{\mathbf{K}}=\sum_i\ (\mathbf{p}_i'-e\mathbf{A}_i'+e\mathbf{B}\times\mathbf{r}_i')+\mathbf{p}_0'+Ze\mathbf{A}_0'-Ze\mathbf{B}\times\mathbf{r}_0' \quad (2)$$

Due to its conservation, the pseudomomentum obeys $[\hat{\mathbf{K}},\mathcal{H}]=0$ and its components commute, i.e., $[\hat{K}_\alpha,\hat{K}_\beta]=0$ for a neutral system. **B** is the magnetic-field vector and M is the total mass of the atom. Using a specific gauge (the symmetric one, see below) it has been shown already several decades ago [14,15] that the pseudomomentum may be associated with the CM motion and may in particular be used to perform a so-called pseudoseparation of the CM motion. Indeed, for vanishing external fields, it becomes identical with the total canonical momentum of the atom. The conservation of the latter allows for a complete separation of the CM and electronic motion in field-free space. In the general form (1) of the Hamiltonian, the vector potential **A** generates the homogeneous magnetic-field $\mathbf{B}=\nabla\times\mathbf{A}$. Without loss of generality, the vector potential can therefore be parametrized as $\mathbf{A}(\mathbf{r}_i')=(1/2)\mathbf{B}\times\mathbf{r}_i'+\nabla'\Lambda(\mathbf{r}_i')$ where Λ is an arbitrary scalar function. As a first step towards a gauge-independent pseudoseparation for multielectron systems, we perform a coordinate transformation from the laboratory frame to the CM frame. Specifically, we introduce the CM of the atom and the relative coordinates of the electrons with respect to the nucleus

$$\mathbf{R}_{at}=\frac{1}{M}\left(m\sum_{i=1}^{N}\mathbf{r}_i'+M_0\mathbf{r}_0'\right);\quad \mathbf{r}_i=\mathbf{r}_i'-\mathbf{r}_0' \quad (3)$$

and the corresponding canonically conjugated momenta $\mathbf{P}_{at},\mathbf{p}_i$, respectively. Applying this change of coordinate frame to the Hamiltonian (1) yields the following transformed Hamiltonian

$$\begin{aligned}\mathcal{H}=&\frac{1}{2m}\sum_{i=1}^{N}\left[\frac{m}{M}\mathbf{P}_{at}+\mathbf{p}_i-\frac{e}{2}\mathbf{B}\times\left(\mathbf{r}_i+\mathbf{R}_{at}-\frac{m}{M}\sum_j\mathbf{r}_j\right)\right.\\&\left.-e\left(\frac{m}{M}\nabla_{R_{at}}+\nabla_i\right)\Lambda_i\right]^2+\frac{1}{2M_0}\left[\frac{M_0}{M}\mathbf{P}_{at}-\sum_i\mathbf{p}_i+\frac{Ze}{2}\mathbf{B}\right.\\&\left.\times\left(\mathbf{R}_{at}-\frac{m}{M}\sum_i\mathbf{r}_i\right)+Ze\left(\frac{M_0}{M}\nabla_{R_{at}}-\sum_i\nabla_i\right)\Lambda_0\right]^2\\&-e\mathbf{E}\sum_i\mathbf{r}_i+V_c(|\mathbf{r}_i-\mathbf{r}_j|;|\mathbf{r}_i|),\end{aligned} \quad (4)$$

where $\nabla_{R_{at}},\nabla_i$ are the derivative operators with respect to the CM and electronic relative coordinates, respectively. Furthermore, $\Lambda_i=\Lambda(\mathbf{r}_i+\mathbf{R}_{at}-(m/M)\Sigma\mathbf{r}_j)$ and $\Lambda_0=\Lambda(\mathbf{R}_{at}-(m/M)\Sigma\mathbf{r}_j)$. The pseudomomentum (2) reads then as follows:

$$\begin{aligned}\hat{\mathbf{K}}=&\mathbf{P}_{at}+\frac{e}{2}\mathbf{B}\times\sum_i\mathbf{r}_i-e\sum_i\left(\frac{m}{M}\nabla_{R_{at}}+\nabla_i\right)\Lambda_i\\&+Ze\left(\frac{M_0}{M}\nabla_{R_{at}}-\sum_i\nabla_i\right)\Lambda_0\\=&\mathbf{P}_{at}+\frac{e}{2}\mathbf{B}\times\sum_i\mathbf{r}_i-e\left(\sum_i\mathbf{\Lambda}_i'-Z\mathbf{\Lambda}_0'\right),\end{aligned} \quad (5)$$

where the prime indicates the derivatives of the scalar function $\nabla\Lambda(\mathbf{r})=\mathbf{\Lambda}'$. The second equality in Eq. (5) arises from the particular dependence of the functions Λ_i and Λ_0 on the arguments $\mathbf{r}_i$ and $\mathbf{R}_{at}$. Since the pseudomomentum is a conserved quantity and since its components commute, it is desirable to construct the common eigenfunctions of the Hamiltonian and the pseudomomentum. To this end, let us decompose the eigenfunctions of the Hamiltonian (4) according to

$$\Psi_{tot}(\{\mathbf{r}_i\};\mathbf{R}_{at})=U(\{\mathbf{r}_i\};\mathbf{R}_{at})\Psi(\{\mathbf{r}_i\}), \quad (6)$$

$$\begin{aligned}U(\{\mathbf{r}_i\};\mathbf{R}_{at})=\exp\left(+i\mathbf{K}\cdot\mathbf{R}_{at}-i\frac{e}{2}\left(\mathbf{B}\times\sum_i\mathbf{r}_i\right)\mathbf{R}_{at}\right.\\\left.+ie\chi(\{\mathbf{r}_i\};\mathbf{R}_{at})\right)\end{aligned} \quad (7)$$

with a so-far unknown function $\chi(\{\mathbf{r}_i\};\mathbf{R}_{at})$. **K** is designed to be an eigenvalue of the operator $\hat{\mathbf{K}}$ (see below). The requirement that $\hat{\mathbf{K}}\Psi_{tot}=\mathbf{K}\Psi_{tot}$ yields the most general expression for the function χ

$$\chi(\{\mathbf{r}_i\};\mathbf{R}_{at})=\left(\sum_i\mathbf{\Lambda}_i-Z\mathbf{\Lambda}_0\right)+f(\{\mathbf{r}_i\}). \quad (8)$$

As a next step, we unitarily transform the Hamiltonian (4) according to $\mathcal{H}\rightarrow U^{-1}\mathcal{H}U$ with U from Eq. (7) together with Eq. (8). After some algebra and rearrangement of the terms involved, we arrive at the Hamiltonian $\mathcal{H}=\mathcal{T}+\mathcal{V}$ with

$$\begin{aligned}\mathcal{T}=&\frac{1}{2m}\sum_i\left(\mathbf{p}_i-\frac{e}{2}\mathbf{B}\times\mathbf{r}_i+e\beta\mathbf{B}\times\sum_i\mathbf{r}_i+e\nabla_i f\right)^2\\&+\frac{1}{2M_0}\left(\sum_i\mathbf{p}_i+e\gamma\mathbf{B}\times\sum_i\mathbf{r}_i+e\sum_i\nabla_i f\right)^2,\end{aligned} \quad (9)$$

$$\mathcal{V}=\frac{1}{2M}\left[\mathbf{K}-Ne\mathbf{B}\times\left(\frac{1}{N}\sum_i\mathbf{r}_i\right)\right]^2-e\mathbf{E}\sum_i\mathbf{r}_i+V_c(\{\mathbf{r}_i\}), \quad (10)$$

where $\beta=m/M$ and $\gamma=Nm-M_0/2M$. The part $\mathcal{T}$ of the Hamiltonian represents the kinetic energy of the electrons in the presence of the magnetic field and this term is, as expected, explicitly gauge dependent via the scalar function f. Indeed, expressing $\mathcal{T}$ in terms of the electronic degrees of freedom (coordinates and velocities) with respect to the nucleus we arrive at

$$\mathcal{T}=\frac{m}{2}\sum_j \dot{\mathbf{r}}_j^2-\frac{m^2}{2M}\left(\sum_j \dot{\mathbf{r}}_j\right)^2, \tag{11}$$

which confirms the above statement that $\mathcal{T}$ is the kinetic energy of the electrons with respect to the nucleus. Furthermore, the final Hamiltonian $\mathcal{H}$ contains the important part $\mathcal{V}$ that is independent of the chosen gauge (no scalar function f occurs in $\mathcal{V}$) and may therefore be interpreted as a a generalized potential. Besides the Coulomb interaction terms V_c and the electric Stark term due to the external electric field the first quadratic term of the potential $\mathcal{V}$ is of particular relevance. Apart from the trivial constant $\mathbf{K}^2/2M$, it gives rise to a motional electric-field term $(e/M)(\mathbf{B}\times\mathbf{K})\Sigma\mathbf{r}_i$ and a diamagnetic term $(e^2/2M)(\mathbf{B}\times\Sigma\mathbf{r}_i)^2$. The relevant quantity occuring in the latter two potential terms is the electronic center of mass (ECM), i.e., $\mathbf{R}=(1/N)\Sigma\mathbf{r}_i$ in the internal coordinate frame. It is therefore the ECM that experiences interactions beyond the Coulomb potential and that enters the generalized potential for multielectron systems. In case of one-electron systems, the above potential reduces to the one derived in [8], and in particular, the ECM reduces to the coordinate vector of the single electron. We remark, that the first quadratic term in Eq. (10), which is according to the above an important part of the total potential $\mathcal{V}$, may be shown to represent the kinetic energy of the CM of the atom. Therefore, the CM kinetic energy of a neutral atom provides a potential for the internal motion of the electrons of the atom. This kinetic energy is due to the vanishing net charge of the system (we are dealing with a neutral atom) independent of any chosen gauge of the external vector potential.

In the following sections, we analyze the above-generalized potential $\mathcal{V}$ and identify the geometrical giant dipole configurations corresponding to singly and multiply excited atoms in crossed fields.

III. GIANT DIPOLE CONFIGURATIONS OF SINGLY EXCITED MULTIELECTRON ATOMS

For one-electron atoms ($N=1$), it is well understood how the decentered giant dipole states arise [8–10]. Above some critical electric-field strength of motional ($\Leftrightarrow$pseudomomentum) and/or external origin, the generalized potential $\mathcal{V}$ in Eq. (10) develops for finite magnetic-field strength simultaneously a saddle point and an outer minimum. Concerning the potential $\mathcal{V}$, three regions have then to be distinguished. Close to the origin, the Coulomb potential is dominating. With increasing distance from the origin, the linear Stark terms become important and cause the appearance of the saddle point. Finally, for large distances, the diamagnetic term of $\mathcal{V}$ becomes significant and an outer minimum evolves. Due to the diamagnetic term, we encounter asymptotically a quadratic confinement perpendicular to the magnetic field. An outer potential well located far from the Coulomb singularity therefore develops and becomes increasingly deeper with increasing electric-field strength [8]. This three-dimensional outer well bears bound states that correspond to highly excited Rydberg states of the atom for which the electron is located far from the proton. As a consequence, these decentered states possess a huge electric dipole moment depending on the absolute value of the electric and magnetic-field strengths.

From a physical point of view, the above scenario may be imagined to exist also for multielectron atoms. Assuming that only one of the electrons is excited in the presence of crossed electric and magnetic fields (for an experimental preparation see [11]) one would then expect that it is captured in an outer well due to the generalized potential in crossed fields. Formally, however, this is less evident since the generalized potential in Eq. (10) acts upon the ECM of the atom and not on a single electron. In the following, we will provide a rigorous background on the existence and properties of giant dipole states for singly excited multielectron atoms.

Having identified the generalized gauge-independent potential $\mathcal{V}$ and the kinetic-energy $\mathcal{T}$, we are now allowed to choose a suitable gauge in order to perform further investigations. For reasons of simplicity, we choose $f=0$ in $\mathcal{T}$. To further prepare the Hamiltonian $\mathcal{H}(f=0)$ in Eqs. (9,10) for an investigation of singly excited giant dipole states, several transformations are required that we shall describe in the following. First, we note that the relative kinetic energy of the electrons with respect to the nucleus contains terms involving the velocities of different electrons [see Eq. (11)]. If we deal with singly excited systems, it is however recommendable to formally separate the excited electron from the electrons of the remaining positively charged core as much as possible. To this end, we perform the coordinate transformation

$$\mathbf{r}_i=\mathbf{r}_i'+\alpha\mathbf{r}_N' \quad \mathbf{r}_N=\mathbf{r}_N' \tag{12}$$

with $\alpha=[m/(N-1)m-M]$. In Eq. (12), the primed and unprimed variables denote the coordinates before and after the transformation, respectively. Starting with the relative coordinates of the electrons with respect to the nucleus Eq. (12) shows that in the coordinate system the coordinates of the Nth electron remain unchanged whereas the coordinates of the remaining $(N-1)$ electrons are now shifted by a small fraction of the position of the Nth electron. This leads to the desired simplification with respect to the kinetic-energy $\mathcal{T}$, which then reads

$$\mathcal{T}=\frac{m(M-m)}{2M}\sum_i^{N-1}\dot{\mathbf{r}}_i^2-\frac{m^2}{M}\sum_{i,j;i<j}^{N-1}(\dot{\mathbf{r}}_i\cdot\dot{\mathbf{r}}_j)+\frac{mM_0}{2(M_0+m)}\dot{\mathbf{r}}_N^2. \tag{13}$$

The Nth electron is now kinetically decoupled from the other $(N-1)$ electrons. The kinetic-energy $\mathcal{T}$ and the generalized potential $\mathcal{V}$ of the new Hamiltonian $\mathcal{H}$ take on the following appearance:

$$\mathcal{T}=\frac{1}{2m}\sum_i^{N-1}\left(\mathbf{p}_i-\frac{e}{2}\mathbf{B}\times\mathbf{r}_i+\frac{em}{M}\mathbf{B}\times\sum_i^{N-1}\mathbf{r}_i+\frac{em}{2(m+M_0)}\mathbf{B}\times\mathbf{r}_N\right)^2+\frac{1}{2(m+M_0)}\times\left(\sum_i^{N-1}\mathbf{p}_i+\frac{e[(N-2)m-M_0]}{2M}\mathbf{B}\times\sum_i^{N-1}\mathbf{r}_i\right.$$

$$+\frac{e(N-1)m}{2(m+M_0)}\mathbf{B}\times\mathbf{r}_N\Bigg)^2+\left(\frac{m+M_0}{2mM_0}\right)$$
$$\times\left(\mathbf{p}_N+\frac{e(Nm^2-M_0^2)}{2(m+M_0)^2}\mathbf{B}\cdot\mathbf{r}_N+\frac{em}{2(m+M_0)}\mathbf{B}\times\sum_i^{N-1}\mathbf{r}_i\right)^2, \tag{14}$$

$$\mathcal{V}=\frac{1}{2M}\left[\mathbf{K}-e\mathbf{B}\times\left(\sum_i\mathbf{r}_i+[1-\alpha(N-1)]\mathbf{r}_N\right)\right]^2$$
$$-e\mathbf{E}\left(\sum_i\mathbf{r}_i+[1-\alpha(N-1)]\mathbf{r}_N\right)+V_c(\{\mathbf{r}_i\},\mathbf{r}_N). \tag{15}$$

The kinetic-energy term for the Nth electron [third quadratic term in Eq. (14)] may be simplified by performing the unitary transformation $U^{-1}\mathcal{H}U$ with

$$U=\exp\left[-\frac{iem}{2(m+M_0)}\left(\mathbf{B}\times\sum_i^{N-1}\mathbf{r}_i\right)\mathbf{r}_N\right], \tag{16}$$

which yields then

$$\mathcal{T}=\frac{1}{2m}\sum_i^{N-1}\Bigg(\mathbf{p}_i-\frac{e}{2}\mathbf{B}\times\mathbf{r}_i+\frac{em}{M}\mathbf{B}\times\sum_i^{N-1}\mathbf{r}_i$$
$$+\frac{em}{(m+M_0)}\mathbf{B}\times\mathbf{r}_N\Bigg)^2+\frac{1}{2(m+M_0)}\Bigg(\sum_i^{N-1}\mathbf{p}_i$$
$$+\frac{e[(N-2)m-M_0]}{2M}\mathbf{B}\times\sum_i^{N-1}\mathbf{r}_i+\frac{e(N-1)m}{(m+M_0)}\mathbf{B}\times\mathbf{r}_N\Bigg)^2$$
$$+\left(\frac{m+M_0}{2mM_0}\right)\left(\mathbf{p}_N+\frac{e(Nm^2-M_0^2)}{2(m+M_0)^2}\mathbf{B}\times\mathbf{r}_N\right)^2. \tag{17}$$

Having separated the kinetic energy of the Nth electron from that of the remaining $N-1$ electrons we are now at an excellent starting point in order to perform an adiabatic separation of the motion of the Nth electron from that of the $N-1$ electrons. This is motivated by the fact that the Nth electron is highly excited and possesses a small kinetic energy and low velocity compared to the remaining $N-1$ electrons that constitute the tightly bound atomic core. Formally, we therefore perform an adiabatic expansion of the total wave function $\Psi(\{\mathbf{r}_i\},\mathbf{r}_N)$, which is an eigenfunction of the Hamiltonian $\mathcal{H}$

$$\Psi(\{\mathbf{r}_i\},\mathbf{r}_N)=\sum_k\psi_k(\{\mathbf{r}_i\};\mathbf{r}_N)\cdot\chi_k(\mathbf{r}_N), \tag{18}$$

where $\psi_k(\{\mathbf{r}_i\};\mathbf{r}_N)$ are the eigenfunctions of

$$H_0'=\mathcal{H}-\mathcal{T}_N \quad \text{with}$$
$$\mathcal{T}_N=\left(\frac{m+M_0}{2mM_0}\right)\left(\mathbf{p}_N+\frac{e(Nm^2-M_0^2)}{2(m+M_0)^2}\mathbf{B}\times\mathbf{r}_N\right)^2, \tag{19}$$

i.e., we have $\mathcal{H}_0'\psi_k(\{\mathbf{r}_i\};\mathbf{r}_N)=\epsilon_k(\mathbf{r}_N)\psi_k(\{\mathbf{r}_i\},\mathbf{r}_N)$. $\mathcal{T}_N$ is the kinetic energy of the excited slow electron. The eigenfunctions ψ_k are therefore solutions to the problem of the interacting core and a static, i.e., spatially fixed external charge at the position $\mathbf{r}_N$ in the presence of the external fields. $\epsilon_k(\mathbf{r}_N)$ are the eigenvalues that depend parametrically on the position $\mathbf{r}_N$ of the Nth electron. We remark that the expansion (18) does not take into account the proper antisymmetrization between the Nth and the $N-1$ electrons. However, exchange effects due to this antisymmetrization are expected to be extremely small due to the large distance between the core and excited electrons. The core Hamiltonian $\mathcal{H}_0'$ may be further simplified by applying the unitary transformation

$$U=\exp\left[\frac{iem}{m+M_0}\left(\mathbf{B}\times\sum_i^{N-1}\mathbf{r}_i\right)\mathbf{r}_N\right], \tag{20}$$

i.e., by transforming $\mathcal{H}_0=U^{-1}\mathcal{H}_0'U$ which yields the significantly simplified core Hamiltonian

$$\mathcal{H}_0=\mathcal{T}_0+\mathcal{V}=\frac{1}{2m}\sum_i^{N-1}\left(\mathbf{p}_i-\frac{e}{2}\mathbf{B}\times\mathbf{r}_i+\frac{em}{M}\mathbf{B}\times\sum_i^{N-1}\mathbf{r}_i\right)^2$$
$$+\frac{1}{2(m+M_0)}\left(\sum_i^{N-1}\mathbf{p}_i+\frac{e((N-2)m-M_0)}{2M}\mathbf{B}\times\sum_i^{N-1}\mathbf{r}_i\right)^2+\mathcal{V}. \tag{21}$$

Inserting the expansion Eq. (18) into the Schrödinger equation $\mathcal{H}\Psi=E\Psi$, projecting on the eigenfunction ψ_l of the core Hamiltonian $\mathcal{H}_0$ and using the orthonormality of the eigenfunctions yields the coupled channel equation of motions for the wave functions of the slow Nth electron

$$[\mathcal{T}_N+\epsilon_l(\mathbf{r}_N)-E]\chi_l=-\sum_k\langle\psi_l|\mathcal{T}_N|\psi_k\rangle\chi_k. \tag{22}$$

Performing an adiabatic approximation means to neglect all nonadiabatic coupling elements $\langle\psi_l|\mathcal{T}_N|\psi_k\rangle$ occuring in Eq. (22). This assumes the validity of the approximation of the true eigenfunction of the total system by a single product of a core eigenfunction ψ_l to $\mathcal{H}_0$ and a solution χ_l to the following equation of motion for the excited Nth electron

$$[\mathcal{T}_N+\epsilon_l(\mathbf{r}_N)-E]\chi_l=0, \tag{23}$$

i.e., we restrict the motion of the slow electron to a single potential-energy surface $\epsilon_l(\mathbf{r}_N)$ created by the fast motion of the core electrons. This potential-energy surface takes on the following appearance:

$$\epsilon_l(\mathbf{r}_N)=\frac{1}{2M}(\mathbf{K}-e\mathbf{B}\times[1-\alpha(N-1)]\mathbf{r}_N)^2 - e\mathbf{E}[1-\alpha(N-1)]\mathbf{r}_N-\frac{e}{M}(\mathbf{B}\times\mathcal{P})(\mathbf{K}-e\mathbf{B}\times[1-\alpha(N-1)]\mathbf{r}_N)-e\mathbf{E}\mathcal{P} + \left\langle \psi_l \left| \mathcal{T}_0+V_c+\frac{e^2}{2M}\left(\mathbf{B}\times\sum_k \mathbf{r}_k\right)^2 \right| \psi_l \right\rangle, \quad (24)$$

where $\mathcal{P}_l=\langle\psi_l|\Sigma\mathbf{r}_i|\psi_l\rangle$ is the electric dipole moment of the electronic state ψ_l in the presence of the crossed fields. The reader should note that the potential $\epsilon_l(\mathbf{r}_N)$ in Eq. (24) depends both explicitly and implicitly via ψ_l on the position $\mathbf{r}_N$ of the excited Nth electron. In the following, we restrict ourselves to the electronic ground state ψ_0 of the atomic core. A careful look at Eq. (24) reveals that the terms depending on $\mathcal{P}$ are negligible compared to, e.g., the first quadratic term. Additionally, the last quadratic term occuring in the expectation value with respect to ψ_0 may safely be neglected. The implicit dependence of the potential energy (24) on $\mathbf{r}_N$ may be obtained by performing a multipole expansion of the Coulomb potential V_c with respect to the outer electron. This is justified by the fact that the excited outer electron is located far from the atomic core, i.e., we have $|\mathbf{r}_N|\gg|\mathbf{r}_i|$. The leading term in this multipole expansion describes the Coulomb interaction of the distant Nth electron with a single positive charge located at the position of the atomic core [see Eq. (25) below]. In other words, the atomic core is represented by a positive net charge. Finally, we arrive at the following good approximation to the potential energy ϵ_0:

$$\epsilon_0(\mathbf{r}_N)\approx\frac{1}{2M}(\mathbf{K}-e\mathbf{B}\times[1-\alpha(N-1)]\mathbf{r}_N)^2 - e\mathbf{E}[1-\alpha(N-1)]\mathbf{r}_N-\frac{e^2}{|\mathbf{r}_N|}+\mathcal{C}, \quad (25)$$

where $\mathcal{C}$ is an irrelevant constant. The Stark-like term due to the external electric-field $\mathbf{E}$ may be combined with the first quadratic term in Eq. (25) by redefining the pseudomomentum $\mathbf{K}'=\mathbf{K}+M\mathbf{v}_d$ where $\mathbf{v}_d$ is the drift velocity of charged particles in the crossed external fields $(\mathbf{E},\mathbf{B})$. Thereby, an additional constant appears that may be included in $\mathcal{C}$, i.e., we have

$$\epsilon_0(\mathbf{r}_N)\approx\frac{1}{2M}(\mathbf{K}'-e\mathbf{B}\times[1-\alpha(N-1)]\mathbf{r}_N)^2-\frac{e^2}{|\mathbf{r}_N|}+\mathcal{C}. \quad (26)$$

Including the term of the next higher order with respect to the multipole expansion would give rise to the interaction of the outer charge with the permanent dipole of the atomic core that results in an interaction term $-e(\mathcal{P}\mathbf{r}_N/|\mathbf{r}_N|^3)$. However, due to the strong decay of this interaction with increasing distance of the outer electron from the remaining atomic core, it is safely assumed to be of very minor importance for the properties of the excited atom. Additionally, it turns out that the adiabatic approximation and the approximations introduced above by neglecting $\mathcal{P}$-dependent terms introduce errors that are of the same order of magnitude as the energies associated with the dipole interaction.

Basing ourselves on Eq. (26), we immediately realize that the resulting equation of motion (23) is very similar to that obtained for the hydrogen atom in Refs. [8]. The corresponding results obtained in these references may therefore be directly transfered to the case of the singly excited multielectron atom investigated here.

Above some critical value of the electric-field strength (of motional or external origin) and correspondingly the absolute value of the pseudomomentum K', an outer well is formed in the potential ϵ_0. This well bears weakly bound states for which the Nth electron is located at a large distance from the remaining positively charged atomic core and therefore these states possess a large electric dipole moment. In analogy to Refs. [8], the condition for the existence of the outer well reads $K'^3>(27/4)MB'$ where $B'=[1-\alpha(N-1)]B$. If we specialize to the situation $\mathbf{B}=(0,0,B),\mathbf{K}'=(0,K',0)$, i.e., the electric field points along the negative x direction, we obtain the position of the saddle point, and in particular, of the minimum of the outer well as solutions to the third-order polynomial $(B'^2/M)x_0^3+(B'K'/M)x_0^2-1=0$. Expanding the potential energy ϵ_0 up to second order around the position of the outer minimum and solving the corresponding equation of motion [see Eq. (23)] for an anisotropic charged oscillator in a magnetic field yields the frequencies

$$\omega_\pm=\frac{1}{\sqrt{2}}((\omega_x^2+\omega_y^2+\omega_c^2)\pm[(\omega_x^2-\omega_y^2)^2+2(\omega_x^2+\omega_y^2)\times\omega_c^2+\omega_c^4]^{1/2})^{1/2};$$

$$\omega_z=\left(\frac{1}{\mu|x_0|^3}\right)^{1/2}, \quad (27)$$

where $\mu=[mM_0/(m+M_0)]$ and

$$\omega_x=\left[\frac{2}{\mu}\left(\frac{B'^2}{2M}+\frac{1}{x_0^3}\right)\right]^{1/2};$$

$$\omega_y=\left[\frac{1}{\mu}\left(\frac{B'^2}{M}-\frac{1}{x_0^3}\right)\right]^{1/2};$$

$$\omega_c=\left(\frac{eB(Nm^2-M_0^2)}{\mu(m+M_0)^2}\right). \quad (28)$$

The spectrum is that of a three-dimensional harmonic oscillator with the above frequencies, i.e., $E_{n_+n_-n_z}=(n_++1/2)\omega_++(n_-+1/2)\omega_-+(n_z+1/2)\omega_z+\mathcal{C}'$. This approximation to the exact spectrum of the giant dipole states is only valid for sufficiently low-energetical excitations. With increasing energy in the outer well, effects due to the anharmonicity of the well become increasingly important and a numerical approach to the solution of the corresponding

equation of motion [Eq. (23) together with Eq. (26)] is necessary [8]. We mention here only that anharmonicity effects are most pronounced in the direction parallel to the magnetic field. This concludes our investigation on the giant dipole states of singly excited multielectron atoms in crossed electric and magnetic fields.

IV. GIANT DIPOLE STATES FOR MULTIPLY EXCITED ATOMS

It is an intriguing perspective to go beyond the previously described singly excited decentered atomic states and to ask the question for the existence and properties of giant dipole configurations for multiply excited atoms. To investigate this problem, let us focus on doubly excited systems, more specifically on two-electron systems (the generalization to arbitrary doubly excited systems is then, according to the above results, straightforward). As a first step, let us introduce a suitable coordinate transformation simplifying the two-electron Hamiltonian (10 for $f=0$) for $N=2$. Since important parts of the generalized potential $\mathcal{V}$ depend only on the ECM and since both electrons are assumed to be highly excited, it is natural to introduce the ECM as a coordinate vector. Additionally, we require that the kinetic energy should become as simple as possible, which leads to the relative vector of the two electrons as a good choice for the second coordinate vector, i.e., we have in total $\mathbf{R}=(\mathbf{r}_1+\mathbf{r}_2)/2$; $\mathbf{r}=\mathbf{r}_1-\mathbf{r}_2$. The transformed Hamiltonian $\mathcal{H}=\mathcal{T}+\mathcal{V}$ therefore decomposes into

$$\mathcal{T}=\frac{1}{2\mu}\left(\mathbf{P}-e\frac{\mu}{\mu'}\mathbf{B}\times\mathbf{R}\right)^2+\frac{1}{m}\left(\mathbf{p}-\frac{e}{4}\mathbf{B}\times\mathbf{r}\right)^2, \quad (29)$$

$$\mathcal{V}=\frac{1}{2M}(\mathbf{K}'-2e\mathbf{B}\times\mathbf{R})^2+\frac{e^2}{|\mathbf{r}|}-Ze^2\left[\frac{1}{\left|\mathbf{R}-\frac{1}{2}\mathbf{r}\right|}+\frac{1}{\left|\mathbf{R}+\frac{1}{2}\mathbf{r}\right|}\right]+\mathcal{C}, \quad (30)$$

where $\mu=2mM_0/M$, $\mu'=2mM_0/M_0-2m$. Analogous to the previous section $\mathbf{K}'$ includes both the motional as well as external electric field [see Eq. (25) and discussion below]. As can be seen from Eqs. (29),(30), our coordinate change decoupled the kinetic-energy terms belonging to the two electronic coordinate vectors and also simplified the field-dependent potential terms in Eq. (30). The above Hamiltonian therefore provides an excellent starting point for further investigations.

In general, we expect that possible doubly excited decenterd configurations correspond to resonances of the atom and these can in particular be yielded by the extrema of the six-dimensional potential $\mathcal{V}(\mathbf{R},\mathbf{r})$ in Eq. (30). We therefore have to find the roots of the six nonlinear coupled equations $\partial\mathcal{V}/\partial\mathbf{r}=0$ and $\partial\mathcal{V}/\partial\mathbf{R}=0$. Without loss of generality, we assume in the following again that the magnetic- and electric-field vectors point along the positive z axis and negative x axis, respectively. A cumbersome calculation yields the following geometrical conditions:

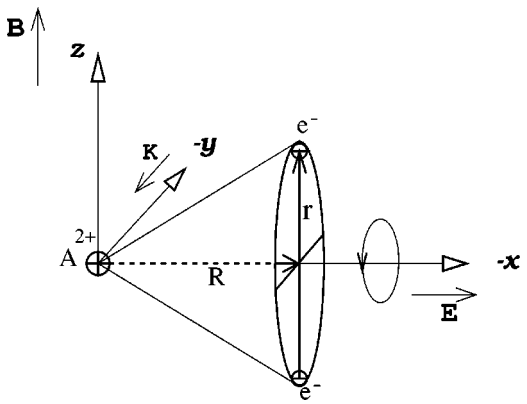

FIG. 1. Shown is a sketch of the geometrical configuration of the giant dipole two-electron resonances. The electric and magnetic-field vector point along the negative x and positive z direction, respectively. **R** is the electronic center of mass coordinate and **r** the relative coordinate vector of the electrons. The big circular loop lying in the yz plane indicates the geometrical position of all extremal configurations.

$$(\mathbf{rR})=0; \quad Y=Z=0; \quad R=\frac{1}{2}\sqrt{3}r;$$

$$P(X)=X^3+\left(\frac{K}{2B}\right)X^2-\frac{3}{8\sqrt{3}}\frac{M}{B^2}=0 \quad (X<0), \quad (31)$$

where $\mathbf{R}=(X,Y,Z)$, $\mathbf{r}=(x,y,z)$ and $r=|\mathbf{r}|$, $R=|\mathbf{R}|$. According to the first geometrical condition $\mathbf{r}\cdot\mathbf{R}=0$, the ECM and the interelectronic coordinate vector are orthogonal. Since $Y=Z=0$, this leads to $x=0$. Furthermore, the condition $R=(1/2)\sqrt{3}r$ and equally $|\mathbf{R}-(1/2)\mathbf{r}|=|\mathbf{R}+(1/2)\mathbf{r}|$ leads to the fact that the two electrons and the nucleus form an equilateral triangle. The remaining nonzero coordinate X of the ECM has to fulfill the corresponding polynomial equation $P(X)=0$ in Eq. (31). This completes the specification of the extremal configurations that are located on a one-dimensional circular manifold. The electrons form a decentered triangular configuration and are *highly correlated* through the fact that they are forced to stay on opposite sides of a circle. The geometry of the extremal configuration described by the above conditions is illustrated in Fig. 1 in which the circular extremal line as well as the opposite electrons are indicated. Both electrons are for laboratory field strengths located far from the nucleus: the electron-nucleus distance scales with $\propto(1/B)$. The position of the extrema will in the following be denoted by $\mathbf{r}_0,\mathbf{R}_0$.

If the inequality $K'^3>(81/4)\sqrt{3}MB$ is fulfilled $P(X)=0$ has two real solutions on the negative x axis (the decentering direction of the atom). The smaller of these two values (excluding the sign) corresponds to a maximum (saddle) of the intersection of the potential $\mathcal{V}$ along the X direction, whereas the larger value yields an outer minimum. Of particular interest is, of course, the case where the ECM is captured in the outer minimum. To investigate the energies and stability of the resonances corresponding to this case we will proceed as follows: We expand the potential $\mathcal{V}$ around the minimum up to second order with respect to all coordinates followed by a normal-mode analysis of the resulting coupled

problem in the presence of the external fields. The expansion of the potential $\mathcal{V}$ reads

$$\mathcal{V}\approx\mathcal{V}(\mathbf{r}_0,\mathbf{R}_0)+\left.\frac{\partial\mathcal{V}}{\partial\mathbf{r}}\right|_{(\mathbf{r}_0,\mathbf{R}_0)}(\mathbf{r}-\mathbf{r}_0)+\left.\frac{\partial\mathcal{V}}{\partial\mathbf{R}}\right|_{(\mathbf{r}_0,\mathbf{R}_0)}(\mathbf{R}-\mathbf{R}_0)+\frac{1}{2}\begin{pmatrix}\mathbf{r}-\mathbf{r}_0\\ \mathbf{R}-\mathbf{R}_0\end{pmatrix}^T\left.\frac{\partial^2\mathcal{V}}{\partial\mathbf{r}\partial\mathbf{R}}\right|_{(\mathbf{r}_0,\mathbf{R}_0)}\begin{pmatrix}\mathbf{r}-\mathbf{r}_0\\ \mathbf{R}-\mathbf{R}_0\end{pmatrix}. \tag{32}$$

Since $\mathbf{r}_0,\mathbf{R}_0$ represents an extremum we have $\partial\mathcal{V}/\partial\mathbf{r}|_{(\mathbf{r}_0,\mathbf{R}_0)}=\partial\mathcal{V}/\partial\mathbf{R}|_{(\mathbf{r}_0,\mathbf{R}_0)}=0$, i.e., the linear terms vanish and the quadratic ones remain only. A tedious calculation gives

$$\left.\left(\frac{\partial^2\mathcal{V}}{\partial\mathbf{r}\partial\mathbf{R}}\right)\right|_{(\mathbf{r}_0,\mathbf{R}_0)}$$

$$=F\times\begin{pmatrix}
-|X_0| & 0 & 0 & 0 & y_0 & z_0\\
0 & \frac{3y_0^2}{4|X_0|} & \frac{3y_0z_0}{4|X_0|} & y_0 & 0 & 0\\
0 & \frac{3y_0z_0}{4|X_0|} & \frac{3z_0^2}{4|X_0|} & z_0 & 0 & 0\\
0 & y_0 & z_0 & \frac{32|X_0|^4}{27\sqrt{3}}\left(\frac{4B^2}{M}-\frac{15\sqrt{3}}{8|X_0|^3}\right) & 0 & 0\\
y_0 & 0 & 0 & 0 & \frac{32|X_0|^4}{27\sqrt{3}}\left[\frac{4B^2}{M}+\left(\frac{3\sqrt{3}}{2|X_0|^3}-\frac{27\sqrt{3}y_0^2}{32|X_0|^5}\right)\right] & -\frac{y_0z_0}{|X_0|}\\
z_0 & 0 & 0 & 0 & -\frac{y_0z_0}{|X_0|} & \frac{32|X_0|^4}{27}\left(\frac{3}{2|X_0|^3}-\frac{27z_0^2}{32|X_0|^5}\right)
\end{pmatrix}, \tag{33}$$

where $F=27\sqrt{3}/32|X_0|^4$ To be specific, let us focus on the case $y=0,z\neq0$. Equation (33) then reads

$$\left.\left(\frac{\partial^2\mathcal{V}}{\partial\mathbf{r}\partial\mathbf{R}}\right)\right|_{(\mathbf{r}_0,\mathbf{R}_0)}=F\times\begin{pmatrix}
-|X_0| & 0 & 0 & 0 & 0 & \frac{2|X_0|}{\sqrt{3}}\\
0 & 0 & 0 & 0 & 0 & 0\\
0 & 0 & |X_0| & \frac{2|X_0|}{\sqrt{3}} & 0 & 0\\
0 & 0 & \frac{2|X_0|}{\sqrt{3}} & D_X & 0 & 0\\
0 & 0 & 0 & 0 & D_Y & 0\\
\frac{2|X|}{\sqrt{3}} & 0 & 0 & 0 & 0 & D_Z
\end{pmatrix}, \tag{34}$$

where

$$D_X=\frac{32|X_0|^4}{27\sqrt{3}}\left(\frac{4B^2}{M}-\frac{15\sqrt{3}}{8|X_0|^3}\right);$$

$$D_Y=\frac{32|X_0|^4}{27\sqrt{3}}\left(\frac{4B^2}{M}+\frac{3\sqrt{3}}{2|X_0|^3}\right);\quad D_Z=\frac{4|X_0|}{9}. \tag{35}$$

Equation (34) contains a column and row of zeros that correspond to the mode of zero frequency: for the total potential, the circle of extremal configurations is associated with a motion possessing a vanishing vibrational frequency. This fact is strictly valid both for the potential $\mathcal{V}$ in Eq. (32) as well as the exact potential $\mathcal{V}$ in Eq. (30). It will also persist for the full equations of motion, i.e., including the kinetic energy in the presence of the external fields (29). As a next step, one has to perform a normal-mode analysis based on the Hamiltonian $\mathcal{H}=\mathcal{T}+\mathcal{V}$ where $\mathcal{V}$ is now taken from Eq. (32) together with Eqs. (34),(35). As a result, one arrives at the following eigenvalue problem for the harmonic frequencies Ω_i, which are the frequencies (energies) of the resonances of the doubly excited atom in crossed fields

$$(-\Omega_i^2)\mathbf{V}_i = F\times \begin{pmatrix} \frac{2|X_0|}{m} & \frac{-iB\Omega_i}{mF} & 0 & 0 & 0 & -\frac{4|X_0|}{m\sqrt{3}} \\ \frac{iB\Omega_i}{mF} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & -\frac{2|X_0|}{m} & -\frac{4|X_0|}{m\sqrt{3}} & 0 & 0 \\ 0 & 0 & -\frac{2|X_0|}{\mu\sqrt{3}} & -\frac{D_X}{\mu} & -\frac{2iB\Omega_i}{\mu' F} & 0 \\ 0 & 0 & 0 & \frac{2iB\Omega_i}{\mu' F} & -\frac{D_Y}{\mu} & 0 \\ -\frac{2|X_0|}{\mu\sqrt{3}} & 0 & 0 & 0 & 0 & -\frac{D_Z}{\mu} \end{pmatrix} \mathbf{V}_i , \tag{36}$$

where $\mathbf{V}_i=(V_1,\ldots,V_6)_i$ are the six-dimensional eigenvectors belonging to the eigenmodes with eigenfrequencies Ω_i of the atom. The reader should note that the matrix on the right hand side (r.h.s.) of Eq. (36) depends explicitly on the frequencies Ω_i, which is due to the appearance of the kinetic-energy $\mathcal{T}$[see Eq. (29)] in the presence of a magnetic field. A closer look at Eq. (36) reveals that the six-dimensional eigenvalue problem reduces to two three-dimensional ones involving exclusively the subspaces $(V_1,V_2,V_6)_i$ and $(V_3,V_4,V_5)_i$, respectively. In the following, we therefore discuss these two subspaces separately.

The subspace $(V_1,V_2,V_6)_i$ contains the above-mentioned mode with zero frequency $\Omega_2=0$, which corresponds to the eigenvector with $V_1=0$, $V_2\neq 0$, $V_6=0$. Starting from the aligned configuration $x_0=y_0=0$, $z_0\neq 0$ (see Fig. 1) this corresponds to an elongation of the interelectronic vector $\mathbf{r}$ tangential to the circle of extremal configurations. The remaining characteristic polynomial in the subspace $(V_1,V_2,V_6)_i$ is quadratic and leads to the following eigenfrequencies:

$$\Omega_i=\left[\frac{1}{2\mu}\left[FD_Z+\frac{\mu}{m^2}B^2-\frac{2\mu}{m}F|X_0|\pm\sqrt{\left(FD_Z-\frac{\mu}{m^2}B^2+\frac{2\mu}{m}F|X_0|\right)^2+\frac{32\mu}{3m}F^2|X_0|^2}\right]\right]^{1/2}, \quad \text{for } i=1,6. \tag{37}$$

The terms of Ω_i involving F originate from the expansion of $\mathcal{V}$, whereas the terms proportional to B^2 come from the kinetic energy, i.e., the field- and velocity-dependent force. The term $(32\mu/3m)F^2|X_0|^2$ is the only one that stems from off-diagonal couplings of the matrix on the r.h.s. of Eq. (36). The analysis of the subspace $(V_3,V_4,V_5)_i$ leads to a characteristic polynomial of sixth order that contains only even powers of Ω_i. It reads as follows:

$$\begin{aligned}(-\mu^2 m)\Omega_i^6+F\Big(&\mu m D_Y+\mu m D_X+2\mu^2|X_0| \\ &+4F^{-1}\frac{\mu^2 m}{\mu'^2}B^2\Big)\Omega_i^4+F^2\Big(-mD_XD_Y-2\mu D_Y|X_0| \\ &-2\mu|X_0|D_X+\frac{8\mu}{3}|X_0|-8\frac{\mu^2}{\mu'^2}B^2F^{-1}|X_0|\Big)\Omega_i^2 \\ &+2F^3\left(|X_0|D_XD_Y-\frac{4}{3}D_YX_0^2\right)=0, \quad \text{for } i=3,4,5, \end{aligned} \tag{38}$$

which reduces to a third-order polynomial equation if Ω_i^2 is introduced as a variable.

The spectrum finally reads $E=\Sigma_{i=1}^6\Omega_i(N_i+\frac{1}{2})+\mathcal{V}(\mathbf{r}_0,\mathbf{R}_0)$, where the frequencies are determined by the above Eqs. (37),(38). Analyzing the frequencies Ω_i, we find that the two largest ones are almost degenerate and are of the order of half the electronic cyclotron frequency $\Omega_e=-eB/m$. The remaining three frequencies contain the motion parallel to the magnetic field that is governed exclusively by the Coulomb interaction and the heavy particle dynamics. They are significantly smaller than Ω_e. The frequencies Ω_i are therefore different by several orders of magnitude and include in particular all possible nuclear and electronic modes of the excited atom in the presence of the external fields (see Fig. 2). Furthermore, it turns out that for typical laboratory field strengths, all frequencies are real. Within our harmonic analysis around these extremal configurations we therefore encounter no decay of the corresponding resonances, which indicates that they should possess a significant life time. Figure 2 shows the dependencies of the five nonzero frequencies on both the electric- as well as the magnetic-field strengths. With increasing magnetic-field strength all frequencies increase. For those frequencies that are associated with the cyclotron motion of the electronic or nuclear degrees of freedom, this behavior is evident. For the

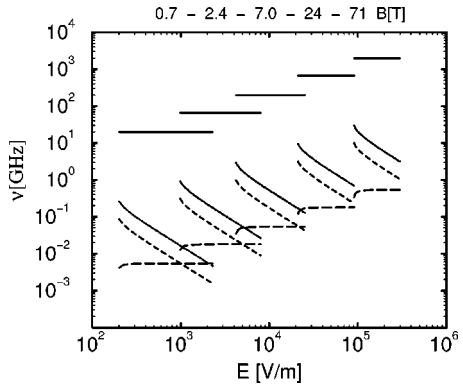

FIG. 2. The five frequencies ν (GHz) of the normal modes belonging to the expansion around the extremal configurations are shown as a function of the applied electric field $E(V/m)$ for different magnetic field strengths. Two of the frequencies are approximately degenerate and indistinguishable (solid line) in the figure: they are associated with the electronic cyclotron motion. The long dashed line shows the behavior of the frequency associated with the cyclotron motion of the heavy particle (nucleus). The dotted and short-dashed line show the behavior of the frequencies associated with the motion along the magnetic field.

frequencies associated with the motion parallel to the magnetic field, it is a consequence of the fact that the position $|X_0|$ of the outer minimum decreases strongly with increasing field strength. The Coulomb potential becomes then stronger and the frequency in the corresponding well parallel to the field raises. As can be seen in Fig. 2, the dependence of the frequencies on the electric-field strength is twofold. Those frequencies associated with the cyclotron motion of the electronic and nuclear degrees of freedom show, in general, only a very weak dependence on the electric-field strength. An exception is the onset of the existence of the outer minimum (well), i.e., $E \approx E_{cr}$ where E_{cr} is the minimal value of the electric-field strength (pseudomomentum) for which the outer-potential well occurs: a strong dependence of the frequency associated with the cyclotron motion of the heavy particle (nucleus) may be observed for a relatively small interval of the electric-field strength. For the frequencies associated with the motion parallel to the magnetic field, we generically obtain a strong dependence on the electric-field strength (see Fig. 2).

The above results address the case of a triangular extremal configuration $y=0$, $z\neq 0$ whose ECM is located at the position $|X_0|$ that corresponds to the outer minimum. In case of the second, i.e., inner extremal configuration, the ECM is located above the saddle in X direction. An analysis similar to the above one reveals then imaginary frequencies describing the decay of the corresponding resonances. The typical lifetimes involved are of the order of nanoseconds. This configuration is certainly of minor interest due to the larger decay rate of the resonances. It is reminiscent of quasi-Penning resonances in crossed fields [16] and a corresponding configuration is of relevance in multiple-ionization processes in strong laser fields [17].

V. CONCLUSIONS

We have established the existence of giant dipole states for highly excited multielectron atoms in crossed electric and magnetic-fields. The basis for our investigation is a gauge-independent pseudoseparation of the CM motion in the presence of the crossed fields. From this pseudoseparation, we obtained a generalized potential whose properties are of central importance to the existence and properties of giant dipole states. In general, we have to distinguish between singly and multiply excited atoms. Using a number of transformations and an adiabatic approximation to separate the fast core electrons from the excited electron we could show for singly excited systems that the properties of their giant dipole states are quite similar to that of the hydrogen atom.

Important facts could be revealed for the giant dipole states of multiply excited atoms. We have shown the existence of a class of highly symmetric giant dipole resonances for doubly excited atoms in crossed fields. For these systems, the electronic CM is captured via the generalized potential. The electrons and the nucleus form an equilateral triangle that possesses a rotational freedom around the axis defined by the nucleus and the electronic CM. This represents a highly correlated electronic and nuclear motion induced by the external fields. The spectrum of this system has been analyzed by performing a harmonic expansion and a subsequent normal-mode analysis. As a result, we obtained complete stability of the resonances up to the second order of the expansion around the equilibrium (extremal) configuration. Of course, a detailed numerical analysis on the above resonances is desirable in order to elucidate the influence of higher-order terms on their properties. The present analytical paper can provide only strong indications towards their existence and characteristics. Also, we have not taken into account the possible process of reorganization of the doubly excited atom to a simply excited one that happens via autoionization. This process is very complicated in the presence of the fields and leads to a singly excited ion that cannot be described in the present framework. However, since the two excited electrons do overlap very little, it is not expected to be very efficient and/or rapid.

It is natural to think of resonances for triply and higher-excited systems. According to the above considerations, we expect the ECM again to be captured by the corresponding potential $\mathcal{V}$ in the outer well. Consequently, the n excited electrons will form a strongly correlated configuration around this decenterd ECM. Some remarks are in order. Due to their spatial extension, quantum-mechanical corrections such as exchange interactions may safely be neglected for the above states. This might be different if we turn to field strengths available in astrophysics, where the size of the multiply excited systems is expected to shrink significantly. Furthermore, the tunneling rates from the outer configuration to the inner Coulomb well may be estimated to be extremely small. To prepare and detect the above resonances in an experimental setup, a scheme similar to the one established in Ref. [11] might be helpful. It relies upon a combined technique of electromagnetic excitation and switching of an external electric field in several steps. The prepared resonances might then be detected via their huge dipole moment.

ACKNOWLEDGMENTS

The author thanks K. Sacha and I. Burghardt for helpful discussions. Part of this work was completed during a visit to the Department of Physics of the University of Nevada at Reno whose kind hospitality is gratefully acknowledged. Financial support by the NSF-DAAD exchange program is acknowledged.

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