

Vibrorotational Structure in Asymmetric Doubly Excited States

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Energy spectra of doubly excited asymmetric (planetary atom) states are found to consist of vibrorotational series of levels. A fully analytical treatment based on approximate adiabatic separation of fast and slow motions allows us to carry out complete analysis and to reveal some substantial differences in the origin of vibrorotational structure in atomic and molecular spectra. [S0031-9007(98)06598-3]

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In 1978 Kellman and Herrick [1] found in the calculated energy spectra of doubly excited states (DES) of the He atom sequences of levels which form vibrorotational progressions

$$E_{KLT} = \omega(n_2 - K - 1) + B[L(L + 1) - T^2]. \quad (1)$$

K was interpreted as vibrational quantum number and L and T as rotational quantum numbers. The underlying model represents an atom as an analog of a symmetric linear three-atomic molecule. In the configuration eZe the electrons reside on opposite sides of the atomic nucleus with the charge Z , being at equal distances from it.

Validity of this model was confirmed by numerous studies of electron density distributions in DES, calculations of expectation values for characteristic operators, and analysis of adiabatic potential curves in hyperspherical coordinates [2–17]. The (K, T) classification of DES became a standard approach. It was realized that vibrorotational spectra in atoms have some unusual features as compared with their analogs in molecules. For instance, they are truncated at some values of total orbital momentum L , exhibit high anharmonicity, and show decrease of effective momentum of inertia $I = 1/2B$ as L increases along rotational series. The understanding of vibrorotational structure is not yet complete. This is testified, for example, by the fact that vibrational frequency ω and rotational constant B are usually obtained by fitting some calculated set of energy levels (see, for instance, Refs. [1,3,6]). Only recently Gerasimovich *et al.* [16] succeeded in deriving some analytical formulas valid for large Z and large $n_1 \sim n_2$.

Vibrorotational structure is manifested most clearly in the intrashell DES with equal principal quantum numbers of electrons n_1 and n_2 , but is discerned also for the intershell states with close n_1 and n_2 . Classification of strongly asymmetric DES (with $n_1 \gg n_2$) has some special features [18,19]. The vibrorotational energy level structure has not been reported for this case.

Recently, considerable attention [20–25] was devoted to the special type of strongly asymmetric DES in which

both electrons reside on the same side of atomic nucleus, being well separated in radial coordinates. In particular, two versions of adiabatic theory for these states [23,24] provide a good agreement with the results of extensive numerical calculations and give important insight into the nature of these *planetary atom states*. However, only S states were considered in the adiabatic approach as well as in the major part of other studies. This type of asymmetric DES have avoided observation even by the most refined experimental technique [26]. (It is difficult to populate these states starting from the low-lying levels.) All of this, of course, did not favor searching for rotational structure.

The present paper extends the adiabatic theory of asymmetric DES to the case of nonzero total orbital momentum L . We find vibrorotational series of levels [see formula (17) below], expose their physical origin, and discuss common features and differences as compared with the molecular vibrorotational spectra. By this study we hope to provide some insight into the more complicated problem of vibrorotational structure in intrashell DES.

The adiabatic approach is developed based on the observation that the inner electron performs *fast motion* over its elliptical orbits. Parameters of the orbit *slowly* evolve in time due to the perturbation by the outer electron, the motion of which is also *slow*. To implement this idea, the Hamiltonian of two-electron atom is first simplified under the assumption that the inner (second) electron has a definite principal quantum number n_2 ,

$$H = -\frac{Z^2}{2n_2^2} + \frac{1}{2} p_{1r}^2 + \frac{l_1^2}{2r_1^2} - \frac{Z}{r_1} + \frac{1}{r_{12}}, \quad (2)$$

where \vec{r}_1 and \vec{r}_2 are electron vectors relative to the atomic nucleus, $\vec{r}_{12} = \vec{r}_1 - \vec{r}_2$, l_1 and l_2 are one-electron orbital momenta, and p_{1r} is the radial momentum for the outer electron. The exchange effects are negligible for the asymmetric states since the electrons are located mainly in nonoverlapping regions of space. Therefore the electrons can be treated as distinguishable particles, with the subscripts 1 and 2 assigned to the outer and inner electron variables, respectively.

The second step involves averaging of (2) over the fast motion,

$$\langle H \rangle = -\frac{Z_{\text{eff}}^2}{2n_2^2} + H'_{\text{rad}} + H_{\text{ang}}^{(0)}, \quad (3)$$

$$H'_{\text{rad}} = \frac{1}{2} p_{1r}^2 + \frac{1}{2} k(r_1 - r_{10})^2, \quad (4)$$

$$H_{\text{ang}}^{(0)} = -\frac{1}{2} k_1 l^2 - \frac{1}{2} k_2 \varphi_{12}^2. \quad (5)$$

Here φ_{12} is the angle between the outer electron vector \vec{r}_1 and the direction of perihelium of the inner electron elliptical orbit (the latter coincides with the time-averaged inner electron vector $\langle \vec{r}_2 \rangle$). The analytical averaging is carried out under the assumption that l_1/n_2 , l_2/n_2 , and φ_{12} are small which leads to the quadratic (harmonic) approximation (4) and (5) [note that the variables l_2 and φ_{12} are implicit in (2)]. The technical details are thoroughly discussed in Ref. [23]. Here we cite only the analytical formulas for the coefficients,

$$Z_{\text{eff}}^2 = Z^2(2 - 3Z^{-2/3} + 2Z^{-1}), \quad \alpha = \frac{2}{Z - Z^{1/3}},$$

$$k = \frac{3Z^{2/3}}{16n_2^6} (Z - Z^{1/3})^4, \quad k_1 = \frac{1}{8n_2^4} (Z - Z^{1/3})^2,$$

$$k_2 = \frac{3Z^{2/3}}{8n_2^2} (Z - Z^{1/3})^2. \quad (6)$$

The first part of the averaged Hamiltonian, H'_{rad} , describes radial oscillations of the outer electron near the equilibrium separation $r_{10} = \alpha n_2^2$. The Hamiltonian $H_{\text{ang}}^{(0)}$ describes oscillations of the angle φ_{12} accompanied by correlated orbital momentum exchange between the electrons. The superscript (0) emphasizes that this expression was obtained for S states which correspond to plane motion with $l_1 = -l_2 \equiv l$ in classical mechanics.

Extending the adiabatic scheme to asymmetric DES with nonzero values of L , we restrict for simplicity our analysis to the states which correspond to the plane classical motion of electrons. In terms of (K, T) classification these are $T = 0$ states. The radial part of the averaged Hamiltonian $\langle H \rangle$ in the simplest approximation is independent of L , being given by Eq. (4). The derivation of the effective angular Hamiltonian is exactly the same as before except for the last step where we had interrelated classical one-electron orbital momenta by the formula $l_1 = -l_2$ valid for S states. For arbitrary L one has to distinguish between l_1^2 and l_2^2 , giving a generalization of (5),

$$H_{\text{ang}}^{(L)} = \frac{l_1^2}{2r_{10}^2} - \frac{1}{2} k_1' l_2^2 - \frac{1}{2} k_2 \varphi_{12}^2. \quad (7)$$

Here the first term is simply a centrifugal potential for the outer electron with the orbital momentum l_1 . The second and the third terms emerge from averaging of the electron-electron interaction operator $1/r_{12}$ over fast inner electron orbital motion under the same assumptions as before. The

coefficients k_1 in (5) and k_1' in (7) are simply related,

$$k_1' = k_1 + \frac{1}{\alpha^2 n_2^4} = \frac{3}{8n_2^4} (Z - Z^{1/3})^2. \quad (8)$$

In order to benefit from conservation of the total orbital momentum L , we change from l_1 and l_2 to the new canonical variables ℓ and \mathcal{Q}

$$l_1 + l_2 = 2\mathcal{Q}, \quad l_1 - l_2 = 2\ell, \quad (9)$$

For the planar motion under consideration one has $\mathcal{Q} = \frac{1}{2}L$. The canonical ‘‘momentum’’ ℓ is conjugate to the canonical ‘‘coordinate’’ φ_{12} , whereas the coordinate conjugate to the momentum \mathcal{Q} does not appear in the Hamiltonian; i.e., it is cyclic.

For S states the variable ℓ oscillates around the stationary point $\ell = 0$. Now we show that for arbitrary L the situation is similar, but for some L -dependent stationary point ℓ_0 . We rewrite (9) in terms of the deviation $\delta\ell \equiv \ell - \ell_0$,

$$l_1 = l_{10} + \delta\ell, \quad l_2 = l_{20} - \delta\ell, \quad (10)$$

$$l_{10} \equiv \ell_0 + \mathcal{Q}, \quad l_{20} \equiv -(\ell_0 - \mathcal{Q}),$$

where l_{10} and l_{20} are the values of one-electron orbital momenta l_1 and l_2 at the stationary point. The term \mathcal{T} in the Hamiltonian (7)

$$\mathcal{T} = \frac{l_1^2}{2r_{10}^2} - k_1' \frac{l_2^2}{2} \quad (11)$$

in the new variables reads

$$\mathcal{T} = \frac{1}{2\alpha^2 n_2^4} (\ell_0 + \mathcal{Q})^2 - \frac{1}{2} k_1' (\ell_0 - \mathcal{Q})^2$$

$$+ \frac{1}{\alpha^2 n_2^4} (\ell_0 + \mathcal{Q}) \delta\ell - k_1' (\ell_0 - \mathcal{Q}) \delta\ell$$

$$+ \frac{1}{2\alpha^2 n_2^4} (\delta\ell)^2 - \frac{1}{2} k_1' (\delta\ell)^2. \quad (12)$$

By imposing the condition that the term linear in $\delta\ell$ vanishes in (12), one obtains an expression for ℓ_0

$$\ell_0 = \frac{1}{k_1} \left(\frac{1}{\alpha^2 n_2^4} + k_1' \right) \mathcal{Q} = \frac{5}{2} L, \quad (13)$$

which does not depend on the nucleus charge Z . At the stationary point we have

$$l_{10} = 3L, \quad l_{20} = -2L, \quad (14)$$

$$\mathcal{T}_0 = \frac{1}{2\alpha^2 n_2^4} l_{10}^2 - \frac{1}{2} k_1' l_{20}^2.$$

Thus the angular part of the effective Hamiltonian $\langle H \rangle$ for nonzero L ,

$$H_{\text{ang}}^{(L)} = BL^2 - \frac{1}{2} k_1 (\delta\ell)^2 - \frac{1}{2} k_2 \varphi_{12}^2, \quad (15)$$

differs from its $L = 0$ counterpart (5) only by the change of variable ($\delta\ell \leftrightarrow l$) and the overall shift by BL^2

with

$$B = \frac{9}{2\alpha^2 n_2^4} - 2k_1' = \frac{3}{8n_2^4} (Z - Z^{1/3})^2. \quad (16)$$

Unexpectedly, B is identical to k_1' (8).

Quantization of harmonic oscillators straightforwardly leads to the serial formula for the asymmetric DES energy levels

$$E_{n_2 n_r n_a L} = -\frac{Z_{\text{eff}}^2}{2n_2^2} + \omega_{\text{rad}} \left(n_r + \frac{1}{2} \right) - \omega_{\text{ang}} (n_a + 1) + BL^2, \quad (17)$$

where n_r and n_a are quantum numbers for oscillators (4) and (15), and the frequencies are

$$\omega_{\text{rad}} = 2\omega_{\text{ang}} = \frac{\sqrt{3}}{4n_2^3} Z^{1/3} (Z - Z^{1/3})^2. \quad (18)$$

In the spirit of semiclassical approximation one replaces L^2 in (17) by $L(L+1)$.

To interpret (17) we compare our atomic system with the three-atom molecule ABC , where, as conventional, only nuclei A, B, C are shown in the notation; the molecular electrons are implicit, being effectively excluded from consideration of vibrational motion by averaging the full Hamiltonian over fast motion of electrons (Born-Oppenheimer approximation). In the same sense we have to denote our *linear* molecule as $Z\langle e_2 \rangle e_1$, where $\langle e_2 \rangle$ is a “quasiparticle” characterized by slow variables of the inner electron (orbital momentum l_2 and direction of perihelium $\langle \vec{r}_2 \rangle$), which remain after averaging over fast inner electron motion.

The first term in the spectral formula (17) is a constant; the second term corresponds to the stretching vibration of $Z\langle e_2 \rangle e_1$ molecule (valence mode). Namely, the outer electron radial coordinate oscillates with the frequency ω_{rad} around the equilibrium separation $r_1 = r_{10}$. Detailed discussion of both these contributions in case of S states [23] remains valid for arbitrary L under consideration.

The third term deserves more comment. It describes the bending vibration of our $Z\langle e_2 \rangle e_1$ molecule. The angle φ_{12} between the outer electron vector \vec{r}_1 and the direction of perihelium of the inner electron elliptical orbit $\langle \vec{r}_2 \rangle$ oscillates with the frequency ω_{ang} around zero value. The canonically conjugate variable (momentum) $\delta\ell$ oscillates around zero with the same frequency and $\frac{1}{2}\pi$ phase shift. As seen from (10), this implies orbital momentum exchange between the electrons, i.e., oscillation of l_1 and l_2 around l_{10} and l_{20} , respectively. These oscillations are shifted in phase by π to satisfy the constraint $l_1 + l_2 = L$ imposed by the total orbital momentum conservation. The “equilibrium” (or stationary point) values l_{10} and l_{20} are linearly shifted (14) as L varies along the rotational series, but the oscillation frequency ω_{ang} (18) remains the same (in the harmonic approximation).

The frequencies of the stretching and bending vibrations are in an integer ratio $\omega_{\text{rad}}/\omega_{\text{ang}} = 2:1$. The implications of this “Fermi resonance” were already discussed [23]; in the refined adiabatic theory [24] the ratio is distorted.

The term BL^2 in (17) is expected to be related to rotation of the “molecule” $Z\langle e_2 \rangle e_1$ as a whole. Within classical mechanics each particle e_1 and $\langle e_2 \rangle$ has moments of inertia I_1 and I_2 , respectively, relative to the infinitely massive center Z . The orbital momenta are expressed as $l_1 = I_1\Omega$, $l_2 = I_2\Omega$, where the frequency of rotation Ω is common for both particles if the system rotates as a whole. Hence the ratio l_1/l_2 is independent of Ω , or of L , for this type of motion. From the expression (14) we see that this is indeed the case, albeit the ratio $l_{10}/l_{20} = -\frac{3}{2}$ has a negative value. This suggests that one of the moments of inertia is negative.

In order to test consistency of this unusual interpretation and to develop it further let us look at how the term $BL^2 = \mathcal{T}_0$ in the energy is formed. According to (14), it contains two contributions. The first one comes from the angular (centrifugal) part of the outer electron kinetic energy and could be written as $\frac{1}{2}l_{10}^2/I_1$ with the momentum of inertia $I_1 = r_{10}^2$. The latter corresponds to the outer electron residing at the distance $r_{10} = \alpha n_2^2$ from the nucleus. This very transparent classical picture allows us, by the way, to find the frequency of classical rotation for L state: $\Omega = l_{10}/I_1 = 3L/r_{10}^2$.

The second term in \mathcal{T}_0 (14), being rewritten as $\frac{1}{2}l_{20}^2/I_2$, corresponds to the negative momentum of inertia $I_2 = -1/k_1'$ of the particle $\langle e_2 \rangle$. It originates from the averaging of the electron-electron interaction $1/r_{12}$ over the fast motion. Note that *no l_2 -dependent contribution comes from the inner electron kinetic energy*. Indeed, the principal quantum number n_2 is presumed to be conserved, and the inner electron unperturbed energy $-Z^2/2n_2^2$ does not depend on l_2 (due to orbital degeneracy of energy levels in Coulomb field). Using the formulas (6) and (8) we find the ratio $I_1/I_2 = -\frac{3}{2}$ in agreement with the discussion above. Negative values of I_2 have a nontrivial dynamic origin. When the “particle” $\langle e_2 \rangle$ is “rotated” around the center Z , it undergoes some changes in internal structure. This is seen, for instance, from the fact that the shape of the inner electron elliptical orbit at the stationary point characterized by the eccentricity $\epsilon = \sqrt{1 - (l_{20}/n_2)^2}$ is changed with increasing L . This is in variance with the conventional image of particle rotation around the center when it is assumed that its inner structure does not depend on the rotational frequency Ω .

The situation for asymmetric states could be compared with that for intrashell DES where, according to Watanabe and Lin [3], the rotational constant B has a “dominating contribution from bielectronic repulsion instead of kinetic energy.” In our case although the contribution from electron-electron interaction is appreciable (and negative), the kinetic energy contribution prevails.

There is yet another notable feature related to the particular character of the particle $\langle e_2 \rangle$. Namely, a conventional linear ABC molecule possesses two different stretching vibrational modes, whereas our $Z\langle e_2 \rangle e_1$ system has only one. The other degree of freedom is “lost” for vibrational motion being extinguished by the averaging

over fast variable. The latter is analogous to electronic motion in *ABC* molecule and produces a nonoscillator quantum number n_2 .

The rotational series (17) are truncated by the obvious "kinematic" constraint $l_{20} \leq (n_2 - 1)$, hence $L \leq \frac{1}{2}(n_2 - 1)$. Moreover, we can anticipate that noticeable distortions in rotational series could appear even for lower values of L . Indeed, the present development essentially uses the harmonic approximation for the bending vibration (as indicated above, the assumption that l_1/n_2 and l_2/n_2 are small is intrinsic for the derivation). The anharmonicity is known to be appreciable for this mode [23]. However, anharmonic corrections are beyond the scope of the present study. We only emphasize that the vibrational quanta are of order $\sim n_2^{-3}$, whereas the rotational quantum is parametrically smaller ($\sim n_2^{-4}$). Of course, there are many other contributions of the same order $\sim n_2^{-4}$, responsible for anharmonicity. In the present study we select only that which is L dependent and thus generates the rotational level structure.

Applicability of the adiabatic approach to vibrational stretching and bending motions is governed by a small value of the ratio $\omega_{\text{rad}}/\omega_{\text{orb}}$ where characteristic orbiting frequency for the inner electron is $\omega_{\text{orb}} = Z^2/n_2^3$. This parameter is as small as 0.075 for $Z = 2$, but increases with Z attaining a value 0.32 for $Z = 5$. This is a manifestation of the well known fact that for large Z the relative role of electron-electron repulsion decreases in comparison with electron-nucleus attraction and hence the correlation effects become less important.

To summarize, we have found vibrorotational structure in the spectra of asymmetric DES. This case is unique since it allows analytical and complete treatment within harmonic approximation. As in the theory of molecular spectra, our approach is based on approximate separation of fast and slow motions in the system. In both cases the difference of time scales alone is responsible for the system stability and regular character of the energy spectrum. Some important features reflect specifics of the atomic system as compared with the molecule, such as negative effective moment of inertia for one of constituent "atoms" and the "loss" of one stretching mode.

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