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Asymmetrical configurations in Coulombic rigid rotators

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We investigate possible asymmetrical configurations of Coulombic three-body systems ($A^{n-}+B^{m+}+B^{m+}$), within Langmuir's rigid-rotator model. The recently found effect by Poirier [Phys. Rev. A **40**, 3498 (1989)] that in the two-electron atoms asymmetrical configurations appear possible for certain charge ratios n/m (in addition to symmetrical ones) has been generalized to constituents of arbitrary masses. Results for realistic cases of the charge ratios 1/2 and 2/3 are presented. It is found that these systems possess the same type of degrees of instabilities as those investigated by Poirier.

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In a recent paper [1] (to be referred to as I), Poirier discussed possible configurations of two-electron rigid-rotator atomic models. The latter was proposed as early as 1921 by Langmuir [2], and was abandoned since the quantum-mechanical theory was set up. This model has been discussed, in addition to a number of other early semiclassical ones, by Percival and Leopold [3], and was the subject of further investigations by Dimitrijević and Grujić [4] (without accounting for the inertial effects). The rigid-rotator model was subsequently rediscovered by Klar [5], who analyzed its stability. Poirier carried out further analysis of the system and found, for certain residual atoms, an electron charge ratio $0.459<Z<1$ unsymmetrical solutions, with an electron distance ratio $\eta=r_1/r_2$ different from 1.

In the same paper, Poirier suggested that the model may turn out to be applicable to doubly excited atoms, with residual core charges reduced to fractions by the screening effect. This situation, however, is hardly feasible, for the potential of $V(r)=q(r)/r$ type (we use atomic units) which accounts for the screening effect (the Thomas-Fermi statistical model, for instance), or the screening of the central body charge in Wannier-type configurations in the continuum [6], cannot be reduced to a pure Coulomb interaction $V=q/r$. On the other hand, systems of the type $A^{n-}+B^{m+}+B^{m+}$, where A and B are atoms, with charge ratio $Z=n/m=1/2$ and 2/3 appear to be good candidates for these unsymmetrical classical configurations.

In this Brief Report we calculate η for a number of realistic triatomic systems, and evaluate a general mass-ratio solution for η. In order to retain a parallel with the two-electron case as much as possible, we shall closely follow Poirier's procedure [1].

We assume that the classical dynamics applies, and write Newtonian equations

$$m_i\frac{d^2\mathbf{r}_i}{dt^2}=\sum_{j\neq i}\varphi_{ij}\mathbf{r}_{ij}\ ,\quad i=1,2,3\ , \tag{1}$$

with interparticle potential functions

$$\varphi_{ij}=C_{ij}/r_{ij}^3\ ,\quad \mathbf{r}_{ij}\equiv\mathbf{r}_i-\mathbf{r}_j\ . \tag{2}$$

In the center-of-mass system, in matrix form Eqs. (1) read

$$\mathbf{M}\frac{d^2\mathbf{R}}{dt^2}=\mathbf{\Phi}\mathbf{R}\ , \tag{3}$$

$$M_{11}=\mu_{12}+\mu_{13}\ ,\quad M_{22}=\mu_{12}+\mu_{23}\ ,$$
$$M_{12}=M_{21}=-\mu_{12}\ , \tag{4a}$$

$$\mu_{ij}=m_im_j/m\ ,\quad m=m_1+m_2+m_3\ , \tag{4b}$$

$$\mathbf{R}(t)=\{\mathbf{r}_{13},\mathbf{r}_{23}\}^T\ , \tag{5}$$

$$\Phi_{11}=\varphi_{12}+\varphi_{13}\ ,\quad \Phi_{22}=\varphi_{12}+\varphi_{23}\ ,$$
$$\Phi_{12}=\Phi_{21}=-\varphi_{12}\ . \tag{6}$$

Solutions within the rigid rotator configuration are sought in the form

$$\mathbf{r}_{ij}(t)=\mathbf{r}_{ij}(\Omega)e^{i\Omega t}\ , \tag{7}$$

with the time-independent factor satisfying

$$\mathbf{\Lambda}\mathbf{R}(\Omega)=0\ , \tag{8}$$

$$\Lambda_{11}=M_{11}\Omega^2+\Phi_{11}\ ,\quad \Lambda_{22}=M_{22}\Omega^2+\Phi_{22}\ ,$$
$$\Lambda_{12}=\Lambda_{21}=M_{12}\Omega^2+\Phi_{12}\ . \tag{9}$$

A corresponding secular equation for the angular frequency yields

$$\Omega'^2,\Omega''^2=\frac{-B\pm(B^2-4AD)^{1/2}}{2A}\ , \tag{10}$$

$$A=m_1m_2m_3/m\ ,$$
$$B=(\mu_{13}+\mu_{23})\varphi_{12}+(\mu_{12}+\mu_{23})\varphi_{13}+(\mu_{12}+\mu_{13})\varphi_{23}\ , \tag{11}$$
$$D=\varphi_{12}\varphi_{13}+\varphi_{12}\varphi_{23}+\varphi_{13}\varphi_{23}\ .$$

One distinguishes two cases.

(a) $\Omega'\neq0$ (nondegenerate solution). One can show for a system with identical particles 1 and 2 ($\mu_{13}=\mu_{23}$, $C_{13}=C_{23}$) that a so-called Wannier configuration arises ($r_{13}=r_{23}$), with two wing particles moving along a circle on the opposite sides of the third one. The general case of (quasi) Wannierian collinear three-body configurations has been studied by Grujić [7], but without accounting for the inertial effects and the system instabilities. We are interested, however, in less trivial solutions.

(b) $\Omega''=0$. It is convenient to write the relative vector as a sum of a fixed and rotating vectors:

$$\mathbf{r}_{13}(t)=\mathbf{f}+\mathbf{r}(t)\ ,\quad \mathbf{f}=\text{const}\ ,\quad \mathbf{f}\cdot\mathbf{r}=0\ , \tag{12}$$

$$\mathbf{r}=\mathbf{C}_1\cos\Omega' t+\mathbf{C}_2\sin\Omega' t\ ,\quad \mathbf{C}_1\mathbf{C}_2=0\ ,\quad C_1=C_2\ . \tag{13}$$

Making use of degeneracy conditions and (8), one obtains [cf. (18) in I]

$$r_{13}^2=\left[\frac{C_{13}}{\varphi_{13}}\right]^{2/3}=f^2+r^2\ , \tag{14a}$$

$$\mathbf{r}_{23}=-\left[\frac{\varphi_{13}}{\varphi_{23}}\right]\mathbf{f}+\frac{(\mu_{12}+\mu_{13})\Omega'^2+\dfrac{\varphi_{13}^2}{\varphi_{13}+\varphi_{23}}}{\mu_{12}\Omega'^2-\dfrac{\varphi_{13}\varphi_{23}}{\varphi_{13}+\varphi_{23}}}\mathbf{r}\ , \tag{14b}$$

$$\begin{aligned} r_{12}^2&=\left[\frac{C_{12}}{\varphi_{12}}\right]^{2/3}\\ &=\left[1+\frac{\varphi_{13}}{\varphi_{23}}\right]^2 f^2+\left[\frac{\mu_{13}\Omega'^2+\varphi_{13}}{\mu_{12}\Omega'^2-\dfrac{\varphi_{13}\varphi_{23}}{\varphi_{13}+\varphi_{23}}}\right]^2 r^2\ . \end{aligned} \tag{14c}$$

Again we restrict ourselves to the case of two identical particles ($\mu_{13}=\mu_{23}, C_{13}=C_{23}$) and distinguish (geometrically) symmetrical situation.

(i) $\varphi_{13}=\varphi_{23}$. In this case, one arrives at a trivial solution

$$\mathbf{r}_{13}(t)=\mathbf{f}+\mathbf{r}\ , \tag{15a}$$

$$\mathbf{r}_{23}(t)=-\mathbf{f}+\mathbf{r}\ , \tag{15b}$$

$$f^2=\left[\frac{C_{12}}{4C_{13}}\right]^{2/3} r_{13}^2\ , \tag{15c}$$

$$r^2=\left[1-\left[\frac{C_{12}}{4C_{13}}\right]^{2/3}\right] r_{13}^2\ . \tag{15d}$$

This is Langmuir's (Klar's in Poirier's designation) model (see, e.g., Fig. 1 in Ref. [4]), with two identical particles moving in phase along two identical parallel circles (a rotating triangle).

(ii) $\varphi_{13}\neq\varphi_{23}$. This is the case of structurally symmetrical, but configurationally unsymmetrical, systems, with two identical particles situated at vortices of a triangle deprived of any symmetry. We note that unsymmetrical configurations may arise within the so-called asynchronous model of helium [8], as a prototype for two light particles moving in the field of a third heavy one. However, Poirier's configuration appears remarkable in that it is a rigid-body configuration, with an unsymmetrical stationary configuration arising from a sort of broken symmetry, rather than from a difference in initial conditions.

Relations (11) now read

$$\begin{aligned} &A=\mu_{13}(2\mu_{12}+\mu_{13})\ ,\quad D=0\ ,\\ &B=-\frac{2\mu_{13}\varphi_{13}\varphi_{23}}{\varphi_{13}+\varphi_{23}}+(\mu_{12}+\mu_{13})(\varphi_{13}+\varphi_{23})\ , \end{aligned} \tag{16}$$

and from (10) one has

$$\Omega'=\omega' r_{13}^{-3/2}\ , \tag{17a}$$

$$\omega'^2=\left[\frac{2\eta^3}{(1+\eta^3)}-\left[1+\frac{1}{\mu}\right](1+\eta^3)\right]\frac{C_{23}}{m_1}\ , \tag{17b}$$

$$\eta\equiv\frac{r_{13}}{r_{23}}\ , \tag{17c}$$

$$\mu\equiv\frac{m_3}{m_1}\ . \tag{17d}$$

Relation (14a) still holds, but with new vector components

$$f^2=C_{13}^{2/3}\frac{\varphi_{23}^{2/3}\phi-\varphi_{13}^{2/3}}{\varphi_{23}^2\phi-\varphi_{13}^2}\varphi_{23}^{4/3}\varphi_{13}^{-2/3}\ , \tag{18}$$

$$\phi_1=\mu_{12}(\varphi_{13}+\varphi_{23})+\mu_{13}\varphi_{23}\ , \tag{18a}$$

$$\phi_2=\mu_{12}(\varphi_{13}+\varphi_{23})^2(\mu_{12}+\mu_{13})+\mu_{13}^2\varphi_{13}\varphi_{23}\ , \tag{18b}$$

$$\phi=\frac{\phi_1^4}{\phi_2^2}\ , \tag{18c}$$

$$r^2=C_{13}^{2/3}\frac{\varphi_{23}^{4/3}-\varphi_{13}^{4/3}}{\varphi_{23}^2\phi-\varphi_{13}^2}\ , \tag{19}$$

and (14b) and (14c) now read

$$r_{23}^2=\left[\frac{\varphi_{13}}{\varphi_{23}}\right]^2 f^2+\phi r^2\ , \tag{20}$$

$$r_{12}^2=\left[1+\frac{\varphi_{13}}{\varphi_{23}}\right]^2 f^2+\mu_{13}^2(\varphi_{23}-\varphi_{13})^2\left[\frac{\phi_1}{\phi_2}\right]^2 r^2\ . \tag{21}$$

From (18) and (21), one has

$$\begin{aligned} &1+2\eta^3+\eta^6+\frac{2\mu}{1+\mu}\eta^2(1+\eta^2)\\ &\qquad-(1+\eta^2+\eta^4)(1+\eta^3)^{2/3}C^{-2/3}=0\ , \end{aligned} \tag{22}$$

$$C=-\frac{C_{13}}{C_{12}}\ , \tag{23}$$

which is to be compared with (24) in I ($C\equiv Z, \eta\equiv r$). Thus for $C\geq 1$ one root is $\eta=1$, and Langmuir's model is recovered.

From (12) and (14b), one has

$$\mathbf{r}_{23}=\sqrt{\phi}\mathbf{r}-\eta^{-3}\mathbf{f}\ , \tag{24}$$

and for angle θ_{12} between $\mathbf{r}_{13}$ and $\mathbf{r}_{23}$ one obtains

$$\cos\theta_{12}=\frac{\eta[1-(1+\mu)\eta+\eta^2]}{(1+\mu)(1+\eta^2+\eta^4)}\ . \tag{25}$$

As can be verified by direct inspection, the left-hand side of (22) appears to be a reflexive function of η, i.e., $F(\eta)\equiv F(1/\eta)$, as it should be, considering the definition of η, (17c). Equation (22) cannot be solved analytically for η, but numerical solutions for a particular choice of C are easy to find. In the limit $\mu\to\infty$, for $C\leq 1$ one obtains Poirier's limit $\eta\to 1, C\to\frac{1}{4}(3/2)^{3/2}(\approx 0.459)$. For a finite

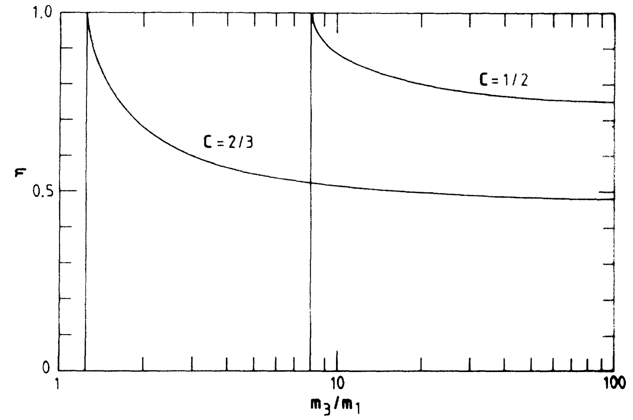

FIG. 1. Distance ratio $\eta = r_{13}/r_{23}$ vs mass ratio $\mu = m_3/m_1$ for two charge ratios $C = -C_{13}/C_{12}$ (see text).

mass ratio, numerical solutions are found for two discrete C values.

(i) $C = 1/2$. In Fig. 1 we plot η against $\mu(=m_3/m_1)$. A minimum mass ratio appears, $\mu_{\min} \approx 8.080$, which allows for unsymmetrical configurations. As μ increases, η slowly tends to its limiting (Poirier's) value $(\sqrt{2}-1)^{1/3}(\approx 0.745)$, attaining at $\mu = 100$ a value $\eta = 0.755$. Note the steep decrease of η just above $\mu_{\min}$.

(ii) $C = 2/3$. This time one obtains $\mu_{\min} \approx 1.25$. Numerical results are also shown in Fig. 1. One notices that for this charge ratio the η curve lies mostly below that for $C = 0.5$, approaching a limiting value somewhat below 0.5 [$\eta(1000) = 0.482$].

We have calculated distance ratios η for a number of realistic atomic systems, as shown in Table I. Halogen atoms are chosen for negative ions because they possess the most stable configurations for an excess of one or two electrons. Thus the electron affinity of negative chlorine is approximately $I(\mathrm{Cl}^-) = 3.07$ eV. We include results for Poirier's system with infinite mass ratio, for the sake of comparison.

Here we ignore the problem of the existence of doubly negative charged ions [9]. We also do not consider the stability of these Coulombic systems, with respect to an electron exchange between oppositely charged ions.

In Fig. 2, we show two examples of asymmetric configurations from Table I. As can be seen from Fig. 2, as one goes from smaller to larger mass and charge ratios, configurations become more symmetric with respect to

TABLE I. Numerical results for asymmetrical configurations of systems [$A^{n-} + 2B^{m+}$].

System	μ	C	η	θ_{12} (deg)	ω' (a.u.)
Poirier's sys.	∞	1/2	0.7454	107.34	1.2872
$\mathrm{I}^- + 2\mathrm{He}^{2+}$	31.964	1/2	0.778 05	106.72	1.5504×10^{-2}
$\mathrm{I}^- + 2^{11}B^{2+}$	11.619	1/2	0.854 66	105.716	9.9603×10^{-3}
$\mathrm{Cl}^- + 2\mathrm{He}^{2+}$	8.8601	1/2	0.9198	105.250	1.7395×10^{-2}
$\mathrm{I}^{2-} + 2^6\mathrm{Li}^{3+}$	21.270	2/3	0.498 62	99.694	2.2878×10^{-2}
$\mathrm{I}^{2-} + 2\mathrm{Kr}^{3+}$	1.5615	2/3	0.771 01	93.371	8.4185×10^{-3}

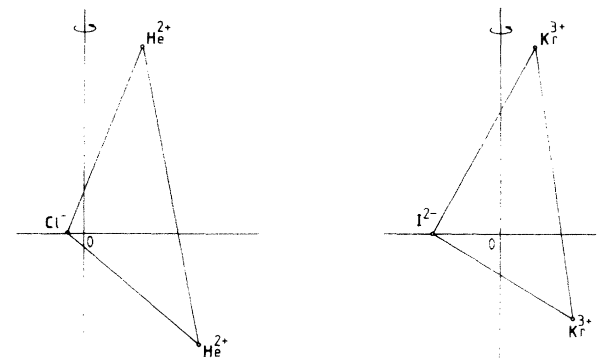

FIG. 2. Rigid-rotator asymmetric configurations for two Coulombic quasimolecular systems.

the horizontal (**r**) axis, and less symmetric with respect to the rotation (**f**) axis.

We now address the problem of dynamic stability of such rotorlike systems, as has been done for the two-electron case [5,6,1]. We write perturbed relative vectors in the cylindrical coordinates:

$$\mathbf{r}_{ij} = (\rho_{ij}^0 + \delta_{\rho ij})\mathbf{e}_\rho + \delta_{\phi ij}\mathbf{e}_\phi + (z_{ij}^0 + \delta_{zij})\mathbf{e}_z \ . \tag{26}$$

After expanding potential functions (2) and retaining linear terms, we substitute it into the equation of motion, written in the form

$$(\mu_{12} + \mu_{i3})\ddot{\mathbf{r}}_{i3} - \mu_{12}\ddot{\mathbf{r}}_{j3} = \varphi_{i3}\mathbf{r}_{i3} + \varphi_{12}\mathbf{r}_{ij} \ , \quad j \neq i = 1,2 \ . \tag{27}$$

Writing deviations in the form

$$\delta(t) = \delta(\lambda)e^{\lambda t} \ , \tag{28}$$

we set down equations for the deviations in matrix form:

$$\mathcal{M}\delta = \mathbf{0} \ , \tag{29}$$

$$\mathcal{M} = \begin{bmatrix} \mathcal{P}_{12} + \mathcal{P}_{13} & -\mathcal{P}_{12} \\ -\mathcal{P}_{12} & \mathcal{P}_{12} + \mathcal{P}_{23} \end{bmatrix} , \tag{30}$$

$$\mathcal{P}_{ij} = \begin{bmatrix} \mu_{ij}\lambda^2 - A_{ij} & -2\mu_{ij}\Omega\lambda & B_{ij} \\ 2\mu_{ij}\Omega\lambda & \mu_{ij}\lambda^2 - C_{ij} & 0 \\ B_{ij} & 0 & \mu_{ij}\lambda^2 - D_{ij} \end{bmatrix} , \tag{31}$$

$$A_{ij} = \mu_{ij}\Omega^2 + \varphi_{ij}^0 \left[1 - 3\frac{\rho_{ij}^{0^2}}{r_{ij}^{0^2}} \right] , \tag{32a}$$

$$B_{ij} = 3\varphi_{ij}^0 \frac{\rho_{ij}^0 z_{ij}^0}{r_{ij}^{0^2}} \ , \tag{32b}$$

$$C_{ij} = \mu_{ij}\Omega^2 + \varphi_{ij}^0 \ , \tag{32c}$$

$$D_{ij} = \varphi_{ij}^0 \left[1 - 3\frac{z_{ij}^{0^2}}{r_{ij}^{0^2}} \right] , \tag{32d}$$

TABLE II. Numerical values for squared Lyapunov exponents.

System	λ_1^2/Ω^2	λ_2^2/Ω^2	$\lambda_{3,4}^2/\Omega^2$	λ_5^2/Ω^2	λ_6^2/Ω^2
Poirier's sys.	-3.09453	-1	$-0.27473\pm i\times 0.17014$	-1.3×10^{-16}	0.64400
I^-+2He^{2+}	-3.20197	-1	$-0.23966\pm i\times 0.14862$	-4.3×10^{-17}	0.68129
$I^-+2\,^{11}B^{2+}$	-3.39326	-1	$-0.17447\pm i\times 0.057094$	-1.7×10^{-15}	0.74220
Cl^-+2He^{2+}	-3.48851	-1	$-0.23929, -0.04156$	-6.1×10^{-16}	0.76937
$I^{2-}+2\,^6Li^{3+}$	-1.96577	-1	$-0.57883\pm i\times 0.17460$	-2.7×10^{-17}	0.12344
$I^{2-}+2Kr^{3+}$	-2.72925	-1	$-0.24546\pm i\times 0.18446$	4.3×10^{-16}	0.22017

and Lyapunov exponents λ are obtained from the secular equation

$$\det\mathcal{M}=0 . \tag{33}$$

Numerical results are shown in Table II.

We note first that the trivial solutions corresponding to the eigenproblem (29) assume corresponding numerical values up to 14 significant figures ($\lambda_2=\pm i\Omega, \lambda_5=0$). The first of these Lyapunov exponents corresponds to the rotational invariance of the system, and the second to the dilatational invariance. All λ^2/Ω^2 with negative real parts indicate a stable (oscillatory) mode. On the other hand, in all cases one squared exponent (λ_6) turns out to be positive, indicating one stable and one unstable mode (for negative and positive signs of the root, respectively). All systems examined thus appear unstable with respect to radial directions, as is the case with asymmetrical systems (both Langmuir's and Wannier's). We note that the degree of instability, as compared with Poirier's case, appears comparable, except for the last two cases in Table II, which possess smaller λ_6^2/Ω^2 values. Evidently, systems with larger C values are less unstable, which is in accordance with Poirier's findings that systems with $Z\to 1$ tend to become stable. Finally, comparing λ_6^2/Ω^2 values in the two last rows, one sees that as the system possesses more equally distributed masses (μ closer to 1), their stability has decreased. This is understandable, for in the case of an infinite mass constituent the system is merely a spectator, whereas with a mass comparable to the masses of the other two particles correlations play a much more prominent role, disturbing the overall stability.

We note in passing that the present asymmetric configurations could not appear in a quarkonium model, even if one disregards the non-Coulombic part of the interquark interaction, because of the mass ratio restriction. Thus for the proton system u(350 MeV,2/3) $+u$(350 MeV,2/3) $+d$(350 MeV,$-1/3$), one has $\mu\approx 1$, $C=1/2$ (e.g., Ref. [10]), whereas we have $\mu_{\min}=9.080$. For the same reason the lightest negative ion H^- cannot enter an asymmetric Coulombic system, since it would involve $\mu<1$.

How realistic the present quasimolecular model is is difficult to estimate at this stage of investigation. In particular, one should first estimate quantum-mechanical probabilities for electron tunneling from negative to positive ions, as the presumably principal mechanism for nonmechanical system instability.

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