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Multipole structure in asymmetrical double Rydberg states

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We have investigated Ba high-l $8snl$ double Rydberg states produced by multistep laser excitation in the presence of a temporarily applied electric field to probe the Coulomb interaction between the two excited electrons. The spectra for the $6s \rightarrow 8s$ excitation with $n-13$ ($l=12$ to 6) show a strong dependence on the orbital momentum of the external electron, due to multipole effects. To explain the $5fn'l'$ resonances appearing in the $n=13$ and 14 $l=6$ spectra, terms higher than the dipole interaction have to be considered in the multipole expansion.

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The study of electron correlation effects in doubly excited states presents a particularly exciting problem in atomic physics. Because of the nonseparability of its Hamiltonian, the three-body problem cannot be solved easily except when the electrons are asymmetrically excited. Then they are located at different radii ($\langle r_1 \rangle \ll \langle r_2 \rangle$) and considered as distinguishable particles with negligible exchange effects. In the case of core penetrating low-l orbits, the spectra are dominated by the properties of the intershell states ($N_1 l_1 n_2 l_2$) inside a non-Coulombic reaction zone. These states are well described outside of it in the framework of the quantum-defect theory [1], where the long-range electronic interactions are in general neglected. This is no longer valid when the electrons are in a planetary situation [2] because of their weaker penetration in the positive doubly charged core. The electron-electron and electron-core interactions may be comparable; recent calculations [3] for a three-body Coulomb system give new insights into the problem.

Electronic correlation studies in highly doubly excited atoms in the different situations schematically described above have been reported previously. These range from the pure three-body Coulomb systems He [4,5] and H^- [6] to non-Coulombic ones involving electronic collision excitation in He^- [7,8] and sequential laser excitation of both electrons in alkaline-earth-metal atoms [9–15]. To overcome the autoionization broadening of the lines, doubly excited states with a non-core-penetrating high-l outer electron were produced by an electric-field switching method [16,17]. The results indicated an n^{-3} dependence and a rapid decrease with l of the autoionization rates due to the centrifugal barrier potential, which prevents penetration of the nl electron inside the residual core. In this case, the outermost electron moves approximately in a hydrogenic orbit around a singly charged core. Then, the anisotropic part of the Coulombic electron-electron interaction can be written, assuming $r_1 < r_2$ as [18]

$$1/r_{12} - 1/r_2 = \sum_{q=1}^{\infty} r_1{}^q r_2{}^{-(q+1)} C^q(\theta_1,\varphi_1) C^q(\theta_2,\varphi_2) , \tag{1}$$

where the $C^q(\theta,\varphi)$ are q rank tensorial operators. The total Hamiltonian, including the spin-orbit interaction of the inner $N_1 l_1$ electron but neglecting that of the outer one, commutes with K^2 and K_z as well as the parity operator. Here K denotes the coupled angular momentum between the total angular momentum j_1 of the $Ba^+(N_1 l_1)$ core and the l momentum of the outer electron in a $(j_1 l)K$ scheme. This model allows us to understand the $6p_{3/2}nl$, $6d_{5/2}nl$ [12], and $7d_{5/2}nl$ [13] spectra where the long-range correlations due to dipole and quadrupole terms give $2j_1+1$ components in a so-called K structure. When $j_1 = \frac{1}{2}$ the two $K = l \pm \frac{1}{2}$ components are degenerate [12], as in $N_1 snl$ states. For $10s$ core excitation, we have shown that the electron-electron interaction is essentially dipolar with $\simeq 15\%$ maximum quadrupolar contribution for $l=10$ [19].

The purpose of this experiment is to excite asymmetrical double Rydberg $8snl$ states where the inner $8s$ electron is Ba^{2+}-core-penetrating and where the outer nl electron neither penetrates the Ba^{2+} core nor overlaps the $8s$ orbital. We also intend to study the electron-electron interaction spectral signatures when they start to influence each other more by decreasing l or in other words to observe for the first time the evolution from a quasiplanetary situation to a more complex three-body system. Here, we point out that studying states with an inner $8s$ electron may be not as appropriate as higher-l_1 less-core-penetrating orbits, but avoids the complex K structure [19], which might possibly mask new phenomena in the spectra.

We have used selective two-step excitation of a $6snk$ Stark state in a static field and the adiabatic switching method to produce a $6snl$ initial Rydberg state. Then, double-Rydberg final states were monitored by recording two-photon excitation spectra in the vicinity of the $6s \rightarrow 8s$ Ba^+ ionic transition. Figure 1 shows typical scans for $6s_{1/2}nl \rightarrow 8s_{1/2}nl$ spectra with $n=13$ and $k=11-7$. The spectra are recorded in our experimental setup with a residual stray field (≤ 1 V/cm) in the Stark switching technique, causing small l mixing to be present in the initial state when firing the core-excitation laser. This has been studied elsewhere in detail [19] and does not interfere with the results presented here. It gives the

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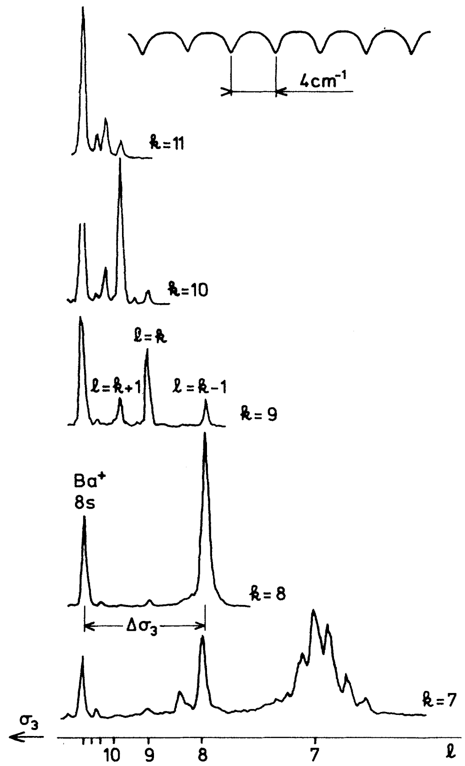

FIG. 1. Two-photon excitation spectra $6s_{1/2}13l \rightarrow 8s_{1/2}13l$ recorded from an initially selected $6s13k$ Stark state with k varying from 11 to 7. Spectra are referred to the $6s_{1/2} \rightarrow 8s_{1/2}$ two-photon Ba$^+$ transition, and the Fabry-Pérot fringe spacing is 4 cm^{-1}. The horizontal scale gives the l value and the position of the identified $8s_{1/2}nl$ lines in the spectra.

presence of weak $l=k\pm1$ components on both sides of the main $l=k$ line, which corresponds to the selected $6snk$ state. It is less pronounced for $k \leq 8$ because the energy splittings between the $6snl$ and $6snl\pm1$ states are larger. The Ba$^+$ $6s \rightarrow 8s$ two-photon transition, on the left side of each spectrum, is derived from the laser populated Ba$^+$ ground-state ions. It gives a useful energy marker for the position of a pure noninteracting two-electron system with the outer one at infinity. The relative energy distance $\Delta\sigma_3$ of each identified $8s_{1/2}nl$ peak from the Ba$^+$ $8s$ transition measures directly the effect of the interaction between the electrons in the final state. This is proportional to the quantum defect difference $\Delta\delta=\delta_{8s}^{13l}-\delta_{6s}^{13l}$ reflecting the change affecting the external nl electron in the presence of the final excited Ba$^+$ state. Notice the increasing value of $\Delta\sigma_3$ as l decreases. This can be understood in a classical picture by comparing the inner turning point of the outer $n=13$ electron, which evolves from 90 a.u. ($l=11$) to 23 a.u. ($l=6$), i.e., close to the outer turning point of the inner $8s$ electron (19 a.u.). Thus the outer electron never penetrates. For $k=7$, we can see that many additional peaks appear in the region of the expected $8s$ $n=13$ $l=7$ single line showing admixture with other doubly excited states. For $l=6$, the phenomenon is more pronounced, as can be seen in Fig. 2 by comparing traces (a) and (b) and characterized by extended structures also present in $n=14$ $l=6$ spectra [trace (c)]. These doubly excited states are identified as belonging to $5f_{5/2,7/2}n'l'$ series because of the regularity of their measured $n^{*\prime}$ effective quantum numbers with respect to Ba$^+$ $5f$ levels [20]. It corresponds to octopole mixing between $8s$ and $5f$ Ba$^+$ core channels in the final state due to multipole effects,

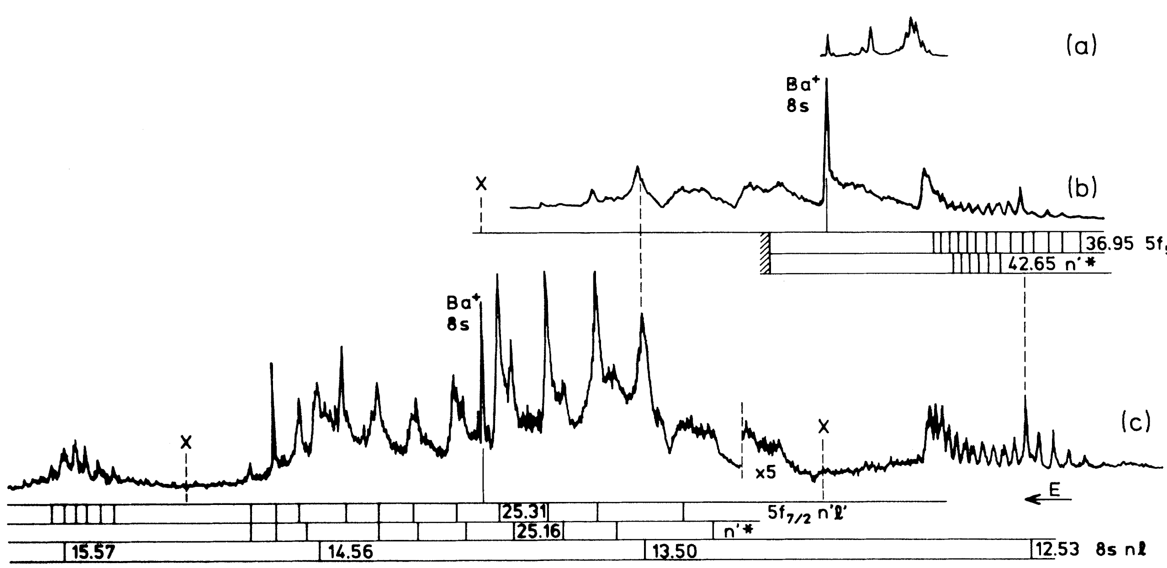

FIG. 2. Two-photon excitation spectra from the $6s_{1/2}nl=6$ initial state with the $n=13$ trace (b) and $n=14$ trace (c) showing the large structure identified with the $5f_{5/2,7/2}n'l'$ series. In order to show the resonances corresponding to the same levels in (b) and (c) both spectra are presented in an absolute energy scale. For this reason the ionic $8s$ Ba$^+$ line is displaced by the energy difference between the two $6sn=13$ and 14 $l=6$ initial levels. The intensity of the $5f_{5/2,7/2}n'l'$ series is modulated by the square of the overlap integral. The $\times$ marks point the zeros of the integral. The trace (a) is identical to the bottom trace $k=7$ of Fig. 1 for $n=13$ and has been reported here for convenience, showing the rapid evolution of the data from $l=7$ to 6.

the importance of which varies inversely with the inner turning point of the outer electron.

Considering the simple perturbative model and applying conservation of parity and K, for octopole coupling with $8s_{1/2}$ $n=13$ $l=6$ ($K=\frac{13}{2},\frac{11}{2}$ degenerate) levels, we predict the $5f_{5/2,7/2}$ $n'l'=9$, 7, 5, or 3 with $K=\frac{13}{2}$ and/or $\frac{11}{2}$ states, giving a total of six K series converging to the $5f_{5/2}$ and eight K series to the $5f_{7/2}$ limits. Assuming a small quantum defect for the high n' and l' outer electron, we can estimate that the lowest observed member of each $5f_{5/2,7/2}$ series has $n'\simeq 40$ and 22, respectively. These fall in a zone where K structures [21] are always unresolved at our laser linewidth (1.5 GHz full width at half maximum). The data should simplify to four $5f_{5/2}$ and four $5f_{7/2}$ series with $l'=9$, 7, 5, or 3. A quantum defect analysis reveals average δ(mod 1) values for only four perturbed series: two converging to $5f_{5/2}$ ($\simeq 0.14$ and $\simeq 0.41$) and two to $5f_{7/2}$ ($\simeq 0.72$ and $\simeq 0.95$) belonging to different available l' momenta of the external electron.

Writing the energy-dependent final-state wave function as $\Psi(E)=\alpha|8sn^*l\rangle+\sum_i\beta_i|5fn_i^{*\prime}l_i'\rangle+$open (and other closed) channels, the intensity of the excitation spectrum can be expressed as

$$\langle 6snl|T^{(2)}|\Psi(E)\rangle^2=|\alpha|^2\langle 6s|T^{(2)}|8s\rangle^2\langle nl|n^*l\rangle^2$$

where $T^{(2)}$ is the two-photon transition dipole operator and n^* is the effective quantum number of the $8snl$ state. The above expression exhibits the well-known square overlap integral [16] intensity envelope observed in our spectra [see trace (c) in Fig. 2], while the $|\alpha|^2$ factor contains the interaction signature between the two electrons.

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[1] M. J. Seaton, Rep. Prog. Phys. **46**, 1 (1983).
[2] I. C. Percival, Proc. R. Soc. London Ser. A **353**, 289 (1977).
[3] K. Richter, J. S. Briggs, D. Wintgen, and E. A. Solov'ev, J. Phys. B **25**, 3929 (1992).
[4] R. P. Madden and K. Codling, Phys. Rev. Lett. **10**, 518 (1963); Astrophys, J. **141**, 364 (1965).
[5] M. Domcke, C. Xue, A. Puschman, T. Mandel, E. Hudson, D. A. Shirley, G. Kaindl, C. H. Greene, H. R. Sadeghpour, and H. Petersen, Phys. Rev. Lett. **66**, 1306 (1991).
[6] P. G. Harris, H. C. Bryant, A. H. Mohagheghi, R. A. Reeder, C. Y. Tang, J. B. Donahue, and C. R. Quick, Phys. Rev. A **42**, 6443 (1990).
[7] S. J. Buckman, P. Hammond, F. H. Read, and G. C. King, J. Phys. B **16**, 4039 (1983).
[8] S. J. Buckman and D. S. Newman, J. Phys. B **20**, L711 (1987).
[9] P. Camus, T. F. Gallagher, J.-M. Lecomte, P. Pillet, L. Pruvost, and J. Boulmer, Phys. Rev. Lett. **62**, 2365 (1989).
[10] U. Eichmann, V. Lange, and W. Sandner, Phys. Rev. Lett. **64**, 274 (1990).
[11] R. R. Jones, Panning Fu, and T. F. Gallagher, Phys. Rev. A **44**, 4260 (1991).
[12] L. Pruvost, P. Camus, J.-M. Lecomte, C. R. Mahon, and P. Pillet, J. Phys. B **22**, 4723 (1991).
[13] P. Camus, J.-M Lecomte, C. R. Mahon, P. Pillet, and L. Pruvost, J. Phys. II France **2**, 715 (1992).
[14] L. Chen, M. Chéret, M. Poirier, F. Roussel, T. Bolzinger, and G. Spiess, J. Phys. II France **2**, 701 (1992).
[15] U. Eichmann, V. Lange, and W. Sandner, Phys. Rev. Lett. **68**, 21 (1992).
[16] W. E. Cooke and T. F. Gallagher, Phys. Rev. Lett. **41**, 1648 (1978).
[17] R. R. Jones and T. F. Gallagher, Phys. Rev. A **38**, 2846 (1988).
[18] B. R. Judd, *Operator Techniques in Atomic Spectroscopy* (McGraw Hill, New York, 1963), p.79.
[19] P. Camus, C. R. Mahon, and L. Pruvost, J. Phys. B **26**, 221 (1993).
[20] C. E. Moore, *Atomic Energy Levels,* Natl. Bur. Stand. Ref. Data Ser., Natl. Bur. Stand. (U.S.) Circ. No. 35 (U.S. GPO, Washington, DC, 1971), Vol. 3.
[21] P. Camus, J.-M. Lecomte, C. R. Mahon, P. Pillet, and L. Pruvost, in *Tenth International Conference on Laser Spectroscopy, Font-Romeu, 1991,* edited by M. Ducloy, E. Giacobino, and G. Camy (World Scientific, Singapore, 1992), p. 431.