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Energies of the $6snf\ ^1F_3$, $^3F_2$, and $^3F_3$ Rydberg states in Ba I

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We have determined accurate values for the energy of the $6snf\ ^1F_3$, $6snf\ ^3F_2$, and $6snf\ ^3F_3$ states of Ba I with principal quantum number n ranging from 13 to 45, using laser—atomic-beam spectroscopy. The experimental level energies for the thus-far-unknown $^1F_3$ Rydberg series compare well with predicted values from hyperfine-structure measurements in the $6snf\ ^3F_{2,3}$ series for $n > 30$ and from Stark-effect studies in the $6snd\ ^1D_2$ series.

The Rydberg states of barium, particularly the even-parity states, have been the subject of many investigations. The energies of the even-parity states with total angular momentum $J \leq 5$ are known.[1–4] In a multichannel quantum-defect theory (MQDT) analysis of the energies of the levels pertaining to the $6sns$, $6snd$, and $6sng$ Rydberg series, Aymar *et al.*[3,5,6] have shown that these series are perturbed by doubly-excited states belonging to the $6pnp$, $5dns$, and $5dnd$ configurations. The influence of these perturbations is also reflected in atomic properties such as lifetime, hyperfine structure, isotope shift, and polarizability (see, e.g., Refs. 7–11).

Less attention has been given to the odd-parity states. The $6snp\ ^1P_1$ states have been observed by Garton and Tomkins[12] and by Armstrong *et al.*[13] The latter group also measured the energies of the $6snp\ ^3P_1$ and $6snp\ ^3P_2$ states and performed an MQDT analysis of their results. The $6snp\ ^1P_1$ series was found to be more strongly perturbed than the $6snp\ ^3P_1$ series. The perturbers belong to the $5dnp$ and $5dnf$ configurations. Only a few states of the $6snp\ ^3P_0$ series are known.[13] A similar situation exists for the $6snf$ states. Carlsten *et al.*[14] determined the positions of the $6snf\ ^3F_{2,3,4}$ states with $n = 4$–30 and Armstrong *et al.*[13] measured the energy of a number of $^3F_2$ states ($n = 9$–12 and 17–25). Except for a weak perturbation at $n = 20$ in the $^3F_2$ series the triplet series appear to be unperturbed above $n = 19$. The hyperfine-structure studies in the $^3F_2$ and $^3F_3$ series for values of n ranging from 19 to 55 of Eliel and Hogervorst[15] corroborate this conclusion. Experimental data on the energies of the $6snf\ ^1F_3$ states for $n > 9$ are scarce. However, information on the unknown $^1F_3$ states has been extracted in two ways. Firstly, from their parametric analysis of the hyperfine structure in the $6snf\ ^3F_{2,3}$ states Eliel and Hogervorst[15] have evaluated the position of the $6snf\ ^1F_3$ states in the interval $n = 19$–55. Secondly, van Leeuwen *et al.*[11] infer from Stark-effect measurements in the $6snd\ ^1D_2$ series the energies of the $6snf\ ^1F_3$ states for $n = 14$–30. In this communication we present the results of direct measurements of the energies of the $6snf\ ^1F_3$, $6snf\ ^3F_2$, and $6snf\ ^3F_3$ states ($n = 13$–45). These measurements form part of an extensive study of the odd-parity states of barium with $J \leq 4$ which is in progress in our laboratory.

Only a brief description of our experimental setup will be given here. More details can be found elsewhere.[15] A collimated atomic beam of barium is orthogonally intersected by the output beam of a narrow-band laser source. The $6snf$ Rydberg levels are excited with uv radiation from the metastable $6s\,5d\ ^3D_2$ state, populated in a discharge running through the nozzle of the ovensystem producing the atomic beam. This uv radiation is produced by intracavity frequency doubling of a cw ring dye laser (Spectra Physics 380D) operating on Rhodamine-590 and Rhodamine-610 in combination with an ADA crystal[16] and a $LiIO_3$ crystal,[17] respectively. The visible output of the laser is used for wavelength determination and calibration purposes. A few millimeters downstream from the excitation region the Rydberg atoms are field ionized and the detached electrons are detected with an electron multiplier. The excitation region has been carefully shielded to ensure field-free excitation. The

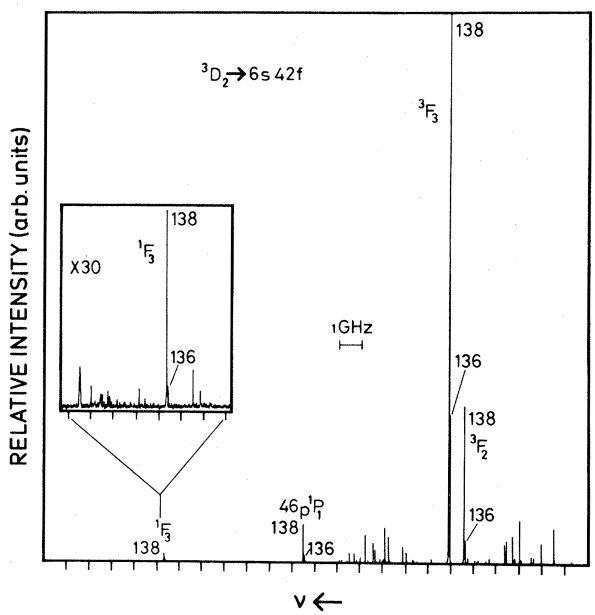

FIG. 1. Excitation spectra $6s\,5d\ ^3D_2 \rightarrow 6s\,42f$. The assignment to the two even isotopes ^{138}Ba and ^{136}Ba is indicated. The remaining peaks are hyperfine components belonging to the odd isotopes ^{135}Ba and ^{137}Ba.

electron-multiplier signal and the signal from a calibration interferometer are stored on a minicomputer.

In Fig. 1 an example of an excitation spectrum $6s\,5d\,^3D_2 \rightarrow 6s\,42f\,^1F_3, ^3F_{2,3}$ is shown. The $6s\,46p\,^1P_1$ level is excited simultaneously. The even isotopes ^{138}Ba (71.7 wt. %) and ^{136}Ba (7.9 wt. %) with nuclear spin $I=0$ both give rise to a single peak for each excitation, whereas the odd isotopes ^{137}Ba (11 wt. %) and ^{135}Ba (6.6 wt. %) with $I=\frac{3}{2}$ display a large number of peaks due to hyperfine interaction.

The energies of the $6snf\,^1F_3$ states ($n=13$–45) and the $6snf\,^3F_{2,3}$ states ($n=13$–45, 47, 50, and 55) have been determined from the transition wavelength for the isotope ^{138}Ba. An evacuated fringe-counting wavemeter, similar to the design of Hall and Lee,[18] is used for this purpose. An iodine-stabilized HeNe laser serves as a reference light source. The wavemeter has been calibrated on the Na D_1 line and no systematic error could be detected on the level of 10^{-4} nm.

The experimental results for transition wavelength and level energy are collected in Table I. For the energy of the $6s\,5d\,^3D_2$ state we have used the value 9215.518 cm^{-1} given by Moore.[19] Three times the statistical error in λ is 5×10^{-5} nm, resulting in an energy uncertainty of 6×10^{-3} cm^{-1}. Our results for the $^3F_2$ and $^3F_3$ states are in agreement with those of Armstrong *et al.*[13] and Carlsten *et al.*,[14] but more accurate.

The results in Table I are represented graphically in Fig. 2. The ordinate is $-n_s^*$ (mod 1), where n_s^* is the effective principal quantum number with respect to the first

TABLE I. Transition wavelength from the $6s\,5d\,^3D_2$ state and level energy for the $6snf\,^1F_3$, $6snf\,^3F_2$, and $6snf\,^3F_3$ Rydberg states.

	$6snf\,^1F_3$		$6snf\,^3F_2$		$6snf\,^3F_3$	
n	λ_{vac} (nm)	E (cm^{-1})	λ_{vac} (nm)	E (cm^{-1})	λ_{vac} (nm)	E (cm^{-1})
13	311.053 99(5)	41 364.277(6)	311.011 34(5)	41 368.685(6)	310.975 41(5)	41 372.402(6)
14	310.168 34(5)	41 456.074(6)	310.120 39(5)	41 461.059(6)	310.097 90(5)	41 463.398(6)
15	309.467 44(5)	41 529.094(6)	309.409 23(5)	41 535.172(6)	309.390 57(5)	41 537.126(6)
16	308.920 87(5)	41 586.264(6)	308.800 14(5)	41 598.922(6)	308.813 91(5)	41 597.480(6)
p[a]	308.570 59(5)	41 623.023(6)				
17	308.301 23(5)	41 651.326(6)	308.334 57(5)	41 647.817(6)	308.345 74(5)	41 646.644(6)
18	307.983 83(5)	41 684.753(6)	307.935 79(5)	41 689.819(6)	307.928 88(5)	41 960.547(6)
19	307.675 97(5)	41 717.244(6)	307.598 79(5)	41 725.398(6)	307.594 27(5)	41 725.879(6)
p[a]	307.413 16(5)	41 745.031(6)				
20	307.182 78(5)	41 769.429(6)	307.314 42(5)	41 755.484(6)	307.308 11(5)	41 756.151(6)
21	306.975 00(5)	41 791.461(6)	307.063 93(5)	41 782.029(6)	307.062 26(5)	41 782.204(6)
22	306.786 57(5)	41 811.471(6)	306.851 00(5)	41 804.625(6)	306.849 42(5)	41 804.795(6)
23	306.616 90(5)	41 829.506(6)	306.665 49(5)	41 824.338(6)	306.664 16(5)	41 824.481(6)
24	306.465 02(5)	41 845.672(6)	306.502 93(5)	41 841.631(6)	306.501 78(5)	41 841.759(6)
25	306.329 26(5)	41 860.132(6)	306.359 79(5)	41 856.876(6)	306.358 77(5)	41 856.988(6)
26	306.207 90(5)	41 873.069(6)	306.233 02(5)	41 870.392(6)	306.232 12(5)	41 870.488(6)
27	306.099 12(5)	41 884.676(6)	306.120 21(5)	41 882.425(6)	306.119 40(5)	41 882.510(6)
28	306.001 50(5)	41 895.096(6)	306.019 35(5)	41 893.190(6)	306.018 68(5)	41 893.260(6)
29	305.913 60(5)	41 904.486(6)	305.928 93(5)	41 902.852(6)	305.928 33(5)	41 902.916(6)
30	305.834 17(5)	41 912.978(6)	305.847 45(5)	41 911.556(6)	305.846 91(5)	41 911.615(6)
31	305.762 14(5)	41 920.678(6)	305.773 81(5)	41 919.431(6)	305.773 33(5)	41 919.480(6)
32	305.696 74(5)	41 927.675(6)	305.707 04(5)	41 926.573(6)	305.706 65(5)	41 926.615(6)
33	305.637 14(5)	41 934.054(6)		41 933.071(6)[b]	305.645 94(5)	41 933.112(6)
34	305.582 70(5)	41 939.884(6)	305.590 89(5)	41 939.006(6)	305.590 55(5)	41 939.043(6)
35	305.532 77(5)	41 945.212(6)	305.540 21(5)	41 944.436(6)	305.539 89(5)	41 944.468(6)
36	305.487 07(5)	41 950.130(6)	305.493 73(5)	41 949.412(6)	305.493 44(5)	41 949.444(6)
37	305.444 95(5)	41 954.642(6)		41 953.995(6)[b]	305.450 72(5)	41 954.025(6)
38	305.406 16(5)	41 958.801(6)		41 958.218(6)[b]		41 958.245(6)[b]
39	305.370 24(5)	41 962.650(6)		41 962.117(6)[b]	305.374 99(5)	41 962.141(6)
40	305.336 99(5)	41 966.216(6)		41 965.721(6)[b]		41 965.745(6)[b]
41	305.306 13(5)	41 969.526(6)	305.310 40(5)	41 969.075(6)	305.310 15(5)	41 969.097(6)
42	305.277 51(5)	41 972.600(6)	305.281 37(5)	41 972.187(6)	305.281 19(5)	41 972.203(6)
43	305.250 74(5)	41 975.470(6)		41 975.081(6)[b]		41 975.103(6)[b]
44	305.225 86(5)	41 978.143(6)		41 977.782(6)[b]		41 977.799(6)[b]
45	305.202 67(5)	41 980.633(6)	305.205 76(5)	41 980.300(6)	305.205 64(5)	41 980.311(b)
47			305.163 19(5)	41 984.869(6)	305.163 04(5)	41 984.885(6)
50				41 990.717(6)[b]	305.108 67(5)	41 990.728(6)
55				41 998.408(6)[b]	305.037 08(5)	41 998.416(6)

[a]States with largest perturber fraction (see text).
[b]Energy derived from the splittings in the spectra.

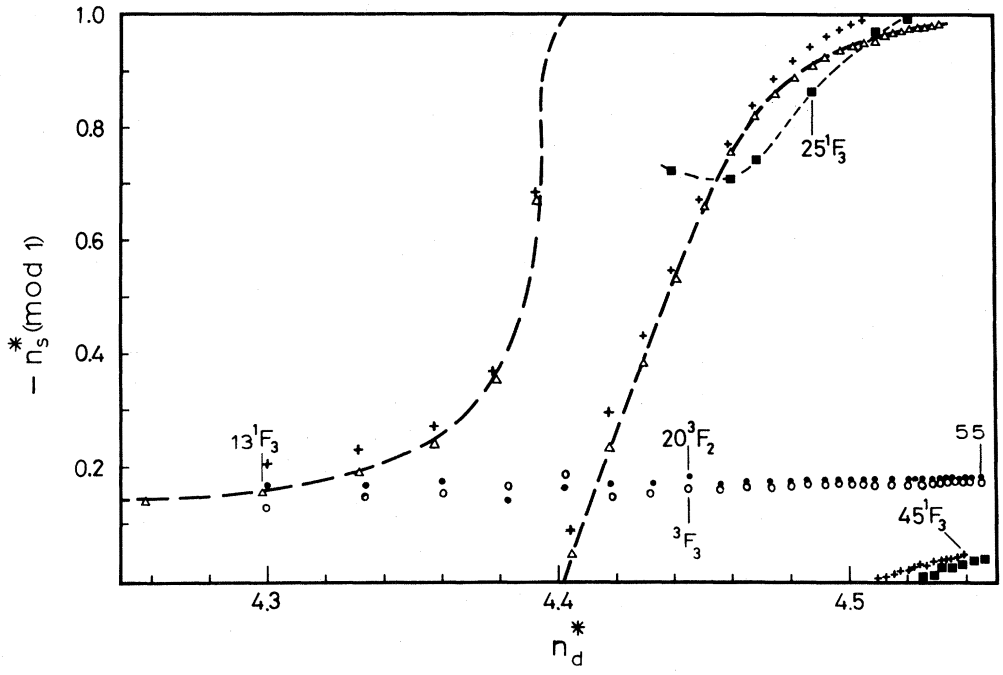

FIG. 2. Partial Lu-Fano plot of $^1F_3$ states (+), $^3F_2$ states (●), and $^3F_3$ states (○). The position of $^1F_3$ states predicted from Stark effect data (Ref. 11) (△) and hyperfine structure (Ref. 5) (■) are included. The curves through the various sets of data points serve to guide the eye.

ionization limit I_s. For this limit we have used $I_s = 42\,034.93$ cm^{-1}. With this value of I_s our recent determination of the $6snp\ ^3P_0$ level energies leads to a constant quantum defect for this series.[20] The effective quantum number n_d^* (with respect to the mean value of the $5d$ limits, 46 908.99 and 47 709.96 cm^{-1}) is the abscissa. The partial Lu-Fano plot clearly demonstrates that the $^1F_3$ series is influenced by two perturbers. The perturbers are assumed to belong to the $5dnl$ series (see Ref. 11). A tentative three-channel MQDT analysis for the $^1F_3$ series based on the experimental energies, taking the average of the two $5d$ limits as the ionization limit for the $5dnl$ series, has been performed to obtain a preliminary classification for the $^1F_3$ series (see Table I). The states with the largest perturber fraction have been labeled with p.

The level positions predicted from the hyperfine-structure analysis of the triplet series[15] and from the Stark-effect data for the $^1D_2$ series[11] are included in Fig. 2. Considering that the predictions from the Stark-effect data, obtained from experimental results in the interval $n = 14$–30, are most reliable for $n \lesssim 30$ and the predictions from the hyperfine-structure analysis are expected to be more accurate for higher values of n (see Ref. 11), the agreement with the present experimental results is satisfactory. This, in particular, demonstrates that Stark-effect data can be applied diagnostically to probe unknown Rydberg series.

The data points for the $^3F_2$ and $^3F_3$ series fall on nearly horizontal lines in the Lu-Fano plot. For the $^3F_2$ series weak perturbations are observed near $n = 16$ and 20. The deviation from the horizontal line in the $^3F_3$ series at $n = 17$ and 18 is due to the crossing of the $^3F_3$ and $^1F_3$ series. Extensive MQDT analyses for the odd parity $J = 2$ and 3 series are in preparation and will be published elsewhere.[20]

The wavelength region in which cw uv laser radiation is available now ($\lambda = 292$–315 nm) also enables the study of the $^3F_4$ states via excitation from the metastable $6s\,5d\ ^3D_3$ state, thus completing the experimental data on the fine structure of the $6snf$ configuration. These data are of importance to test the validity of the parametric analysis of the hyperfine structure in the $6snf$ configuration by Eliel and Hogervorst,[15] who introduced a parameter representing an effective two-body spin-orbit interaction to get good agreement with experimental results. This parametric analysis is strictly valid only if the $6snf$ states are unperturbed. The results of van Leeuwen *et al.*[11] and of the present experiment, however, show this assumption to be incorrect for at least the lower ($n < 30$) $^1F_3$ states.

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[1]D. J. Bradley, P. Ewart, J. V. Nicholas, and J. R. D. Shaw, J. Phys. B 6, 594 (1973).
[2]J. R. Rubbmark, S. A. Borgström, and K. Bockasten, J. Phys. B 10, 421 (1977).
[3]M. Aymar, P. Camus, M. Dieulin, and C. Morillon, Phys. Rev. A 18, 2173 (1978).
[4]P. Camus, M. Dieulin, and A. El Himdy, Phys. Rev. A 26, 379 (1982).
[5]M. Aymar and O. Robaux, J. Phys. B 12, 531 (1979).
[6]M. Aymar and P. Camus, Phys. Rev. A 28, 850 (1983).
[7]P. Grafström, C. Levinson, H. Lundberg, S. Svanberg, P. Grundevik, L. Nilsson, and M. Aymar, Z. Phys. A 308, 95 (1982).
[8]W. Hogervorst and E. R. Eliel, Z. Phys. A 310, 19 (1983).
[9]H. Rinneberg and J. Neukammer, Phys. Rev. A 27, 1779 (1983).
[10]E. R. Eliel and W. Hogervorst, J. Phys. B 16, 1881 (1983).
[11]K. A. H. van Leeuwen, W. Hogervorst, and B. H. Post, Phys. Rev. A 28, 1901 (1983).
[12]W. R. S. Garton and E. S. Tomkins, Astrophys. J. 158, 1219 (1969).
[13]J. A. Armstrong, J. J. Wynne, and P. Esherick, J. Opt. Soc. Am. 69, 211 (1979).
[14]J. L. Carlsten, T. J. McIlrath, and W. H. Parkinson, J. Phys. B 8, 38 (1975).
[15]E. R. Eliel and W. Hogervorst, Phys. Rev. A 27, 2995 (1983).
[16]E. R. Eliel, W. Hogervorst, K. A. H. van Leeuwen, and B. H. Post, Opt. Commun. 39, 41 (1981).
[17]W. A. Majewski, Opt. Commun. 45, 201 (1983).
[18]J. L. Hall and S. A. Lee, Appl. Phys. Lett. 29, 367 (1976).
[19]C. E. Moore, *Atomic Energy Levels,* Natl. Stand. Ref. Data Ser., Natl. Bur. Stand. (U.S.) Circ. No. 35 (U.S. GPO, Washington, DC, 1971), Vol. III.
[20]B. H. Post, W. Vassen, and W. Hogervorst (unpublished).