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Rinneberg, H; Neukammer, J; Jonsson, G; Hieronymus, H; Konig, A; Vietzke, K

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High-n Rydberg Atoms and External Fields

H. Rinneberg, J. Neukammer, G. Jönsson,[a] H. Hieronymus, A. König, and K. Vietzke
Institut für Atom- und Festkörperphysik, Freie Universität Berlin, 1000 Berlin 33, Federal Republic of Germany
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We have observed $6snd\ ^1D_2$ Rydberg states of Ba with principal quantum numbers up to $n = 290$. To our knowledge these are the largest atoms which have been produced under laboratory conditions up to now. Rydberg atoms with $n = 78$ were subject to small electric ($F \lesssim 1$ V/cm) and magnetic ($H \lesssim 850$ G) fields to produce highly resolved spectra of Stark and diamagnetic manifolds. Because of interference effects caused by the presence of nonhydrogenic states within the manifolds, the distribution of oscillator strength varies dramatically and was found to be strikingly similar for Stark and diamagnetic manifolds.

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During the last decade, Rydberg atoms have been studied extensively because of their unusual properties.[1] Because of their large size, these atoms are particularly sensitive to external electric and magnetic fields.[2] For example, the quadratic Stark effect scales according to $(n^*)^7F^2$, n^* being the effective principal quantum number. Rydberg states of Cs and Sr have been observed in a vapor cell for principal quantum numbers n up to 140 and 200, respectively.[3,4] In outer space, however, Rydberg states of hydrogen with $n = 390$ have been found.[1] Fabre *et al.*[5] probed the atomic dimension of Rydberg atoms ($24 \leq n \leq 65$) by measuring their transmission through a micrometer-size slit. In this Letter we report the excitation and detection of atomic Rydberg states of Ba with principal quantum numbers up to $n = 290$, corresponding to a diameter of about 0.01 mm. Starting from the atomic ground state, the Rydberg states were reached by resonant two-step excitation via the $6s6p\ ^1P_1$ intermediate level, employing narrow-band cw laser radiation. At lower principal quantum numbers ($70 \leq n \leq 160$), we have studied Rydberg atoms in external electric and magnetic fields. At $n = 78$, for example, fields of $F \lesssim 1$ V/cm and $H \lesssim 850$ G cause extensive l mixing. Previously, Rydberg states of alkali metals have been studied in external electric[6,7] and magnetic[3,8] fields, up to considerably higher field strengths. However, the spectral resolution of our experiment ($\Delta\nu \simeq 5$ MHz) has never been achieved before. We have recorded highly resolved spectra, exhibiting complete Stark and diamagnetic manifolds. The distribution of oscillator strengths within a manifold varies dramatically when a nonhydrogenic state with large quantum defect is shifted into the manifold by the external field. The intensity distributions of Stark and diamagnetic manifolds were found to be strikingly similar, exhibiting a minimum close to the position of the nonhydrogenic state. The minimum is interpreted as a destructive interference of the s and d components of the transition moments involving the intermediate ($6s6p\ ^1P_1$) and final states. This destructive interference is reminiscent of the minimum observed for Fano-Beutler profiles of autoionizing resonances.

The redistribution of oscillator strengths within a Stark manifold due to the presence of a nonhydrogenic state has been discussed qualitatively by Chardonnet *et al.*[9] in terms of the interaction of one discrete state with a quasicontinuum of discrete states. Experimental investigations of the Stark manifolds of cesium by the same authors[9] were hampered by insufficient resolution, precluding the observation of individual Stark components, and only broad signals were recorded. A well-resolved Stark manifold ($m_j = 0$) of sodium at $n = 29$ was reported by Fabre *et al.*,[10] exhibiting a minimum in intensity close to the position of the nonhydrogenic (d) state. However, because of fine structure the spectrum consists of a superposition of predominantly $m_l = 0$ and $|m_l| = 1$ Stark components, which are degenerate within experimental linewidth. The observed intensity distribution was not discussed, neither qualitatively nor quantitatively. In our case, however, the effect of spin can be neglected (i.e., $m_j = m_l$), since the intermediate ($6s6p\ ^1P_1$) as well as final states are known to be singlets to a good approximation. Hence our spectra reveal the interference effects unambiguously and are amenable to a comparison with calculated intensities.

We used two tunable cw dye lasers (see Fig. 1) to populate $6snl$ Rydberg states in an atomic beam by resonant two-step excitation via the $6s6p\ ^1P_1$ intermediate level. The bandwidths of both lasers were about 1 MHz, and we used powers of 200 μW and 30 mW for the first and second transitions, respectively. The Rydberg atoms were ionized predominantly by forward collisions with Ba atoms in the intermediate or metastable $5d6s\ ^1D_2$ states, i.e., by Penning ionization.[11] The Ba^+ ions were detected by means of a quadrupole mass spectrometer (cf. Fig. 1). Counting rates up to 10^6/sec were obtained. Spectra were recorded by scanning of the frequency of the second laser ($\lambda_2 \simeq 417.5$ nm) while the wavelength of the first laser was stabilized to the resonance line of ^{138}Ba ($\lambda_1 = 553.7$ nm). A parallel-plate capacitor served to compensate stray electric fields or to apply electric fields along the atomic beam axis. Furthermore, two coils taken from a Varian electromagnet were used in a

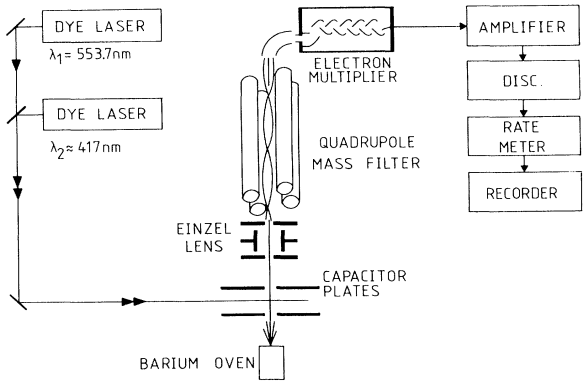

FIG. 1. Experimental setup.

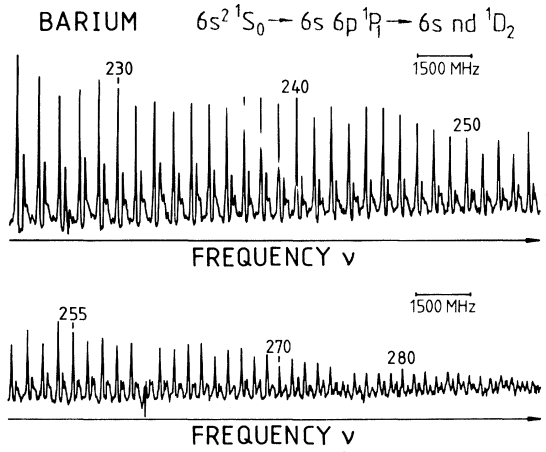

FIG. 2. High-resolution spectrum of barium Rydberg states. Along the vertical axis, intensities are given in arbitrary units. The energy of the $6s\,230d\,{}^1D_2$ state amounts to 42 032.777(2) cm^{-1}. Besides the ${}^1D_2$ series (strong signals), ${}^1P_1$ states appear as smaller lines.

Helmholtz-type arrangement to produce magnetic fields up to 850 G. Since the magnetic field was oriented along the atomic beam, motional Stark effects[2] were suppressed.

Figure 2 shows a high-resolution spectrum of the $6snd\,{}^1D_2$ Rydberg series approaching the ionization limit as close as 40 GHz. The states have been identified by measurement of term values of $6snd\,{}^1D_2$ Rydberg states between $n = 30$ and 290 with an accuracy of ± 60 MHz,[12] with a high-precision Michelson interferometer.[13] From these measurements, we have determined the first ionization limit of Ba to be 42 034.902(3) cm^{-1}. Compared to the ${}^1D_2$ series, the oscillator strengths of $6sns\,{}^1S_0$ states are considerably smaller and hence the ${}^1S_0$ series was followed up to $n = 200$ only. However, because of Stark mixing with neighboring ${}^1D_2$ states caused by stray electric fields, we observe additional signals corresponding to $6snp\,{}^1P_1$ states (cf. Fig. 2). As a matter of fact, in our experiment we are presently limited by stray electric fields of the order of a few millivolts per centimeter, which cause extensive Stark mixing and broadening at the highest principal quantum numbers. By reduction of these fields still further, the gap between measurements performed in the laboratory and observations of Rydberg atoms in outer space by radio astronomy could be closed.

In the remaining part of this Letter we discuss the behavior of high-lying barium Rydberg states in external electric and magnetic fields. We have recorded Stark spectra for principal quantum numbers ranging between $n = 70$ and 160. As an example, we show the linear Stark effect of the hydrogenlike $n = 78$ manifold appearing at the center of Fig. 3(a). At $F = 0$ this manifold corresponds to Rydberg states with angular momentum quantum numbers l ranging through $5 \leq l \leq 77$. In Fig. 3(a) the Stark components of the hydrogenlike manifold are labeled by $k = n_1 - n_2$, where n_1 and n_2 are the parabolic quantum numbers. The electric field was set to $F = 200$ mV/cm, well below the critical field $F_c \approx 9$ V/cm. The splitting $\Delta E = 66$ MHz between neighboring Stark components [cf. Fig. 3(a)] agrees well with the value $\Delta E = 3nea_0F$ obtained from first-order perturbation theory. The transition from the intermediate $6s\,6p\,{}^1P_1$ state to the Stark components probes the s and d character contained in the corresponding wave functions. As can be seen from Fig. 3(a), the intensity distribution and hence the distribution of oscillator strengths is symmetrical to the $k = \pm 1$ components.

It should be noted that the linewidth increases strongly with $|k|$. We tentatively attribute the change in linewidth to small field inhomogeneities due to the center holes in both capacitor plates. Stray electric fields, on the other hand, have no significant influence at $n = 78$. In contrast to the hydrogenlike Stark manifold, barium Rydberg states with angular momentum quantum numbers $l \leq 4$ have quantum defects δ_l sufficiently different from zero[12] and hence exhibit a quadratic Stark effect at $F = 200$ mV/cm. Although Stark mixed, in Fig. 3(a), these states have been labeled according to their LS designation in zero field.

By increasing the electric field strength, we observed dramatic changes in the distribution of oscillator strengths across the Stark manifold. Figure 3(b) displays the spectrum obtained at $F = 400$ mV/cm. The total splitting observed in Fig. 3(b) amounts to about 10 GHz, well below the separation (15 GHz) between neighboring manifolds. The pronounced minimum in intensity appears close to the position to which the $6s\,82s\,{}^1S_0$ state is shifted by second-order Stark effect. This minimum has been observed in the $m_l = 0$ spectrum only. Therefore, we attribute this behavior to an s-d interference of the transition moments between the intermediate ($6s\,6p\,{}^1P_1$) and final (Stark) states. There is little doubt that the experi-

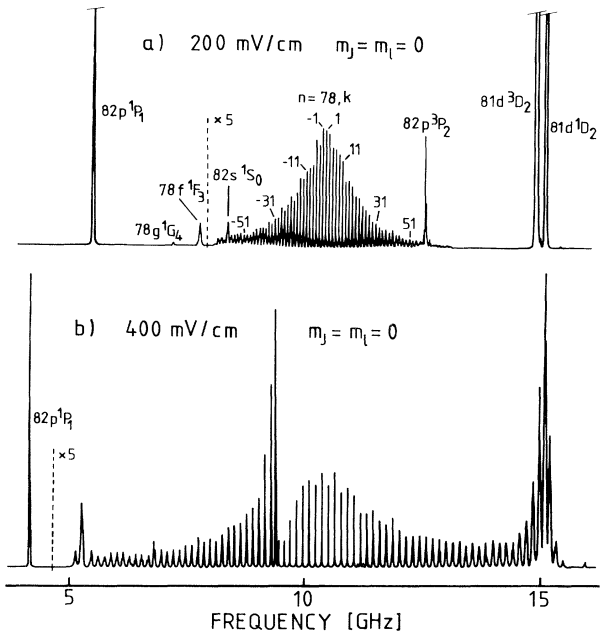

FIG. 3. Stark effect of the hydrogenlike $n = 78$ manifold of Ba. Intensities (vertical axis) are given in arbitrary units. The origin of the frequency scale corresponds to the $6s80d\,^1D_2$ state in zero field [$E = 42016.527(2)$ cm^{-1}]. (a) The Stark components of the hydrogenlike manifold ($\delta = 0$) are labeled by $k = n_1 - n_2$. States with low angular-momentum quantum number ($l \leq 4$) and nonzero quantum defect ($\delta \neq 0$) are characterized by their LS designation in zero field. (b) Interference of the $6s82s\,^1S_0$ (center) and $6s81s\,^{1,3}D_2$ states with the hydrogenlike $n = 78$ Stark manifold.

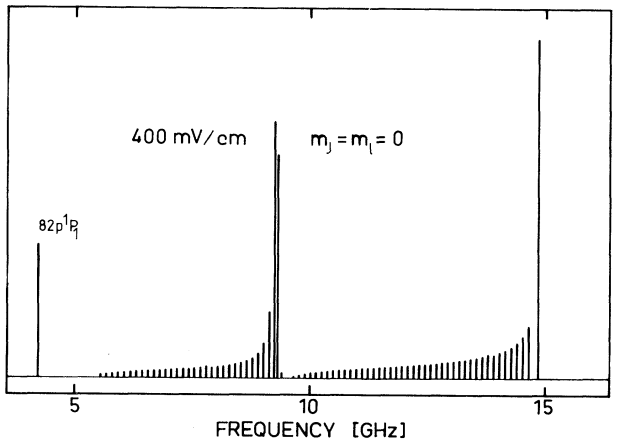

FIG. 4. Calculated distribution of relative oscillator strengths within the $n = 78$ Stark manifold [cf. Fig. 3(b)].

mentally observed splittings as well as the relative intensities could be accounted for quantitatively by diagonalization of the Stark Hamiltonian using a sufficiently large spherical basis set.[6] However, such a procedure is extremely unwieldy in the range of principal quantum numbers under discussion. A superior approach has recently been suggested by Harmin[14] to account for the Stark effect of nonhydrogenic atoms. Starting from the true Stark wave functions of hydrogen and including quantum defects, the theory provides energies and oscillator strengths without diagonalization. The theory has recently been applied[15] to alkali metals at $n = 15$, and good agreement with experiment[6] was achieved. It should be noted that the theory in its present form requires a nonexcited spherically symmetric core. Hence it does not include perturbations due to doubly excited configurations, well known to perturb the Rydberg series of barium. Nevertheless, the admixture of the doubly excited configurations into the wave functions is very small and nearly constant, if we restrict ourselves to a sufficiently narrow energy range. With the $82s\,^1S_0$, $82p\,^1P_1$, and $81d\,^1D_2$ Rydberg states taken to be the only nonhydrogenic states within the energy range under discussion, the intensity distribution of the $n = 78$ Stark multiplet has been calculated following the procedure outlined in Sect. IIIA of Ref. 15. Quantum defects and the relative oscillator strengths of the $6s6p\,^1P_1 \rightarrow 6s82s\,^1S_0, 6s81d\,^1D_2$ transitions in zero field were taken from experiment. With the exception of the $6s82p\,^1P_1$ state, calculated (cf. Fig. 4) and experimental [cf. Fig. 3(b)] relative intensities agree within a factor of 2, approximately. It should be noted that the calculated distribution of oscillator strengths corresponds to constant linewidth, whereas experimentally the linewidth increases towards the lower and upper boundaries of the manifold. This causes Stark components at the center of the manifold to exhibit correspondingly enhanced experimental amplitudes. Although quantitative differences remain between calculated and experimental intensities, the minimum near the center caused by the s-d interference is well reproduced.

In order to explore this interference for the case of a nonhydrogenic state embedded in a diamagnetic manifold, we have applied external magnetic fields to barium Rydberg states within the same energy range. The spectrum shown in Fig. 5 demonstrates l mixing of the $n = 78$ hydrogenlike manifold. Again, the total splitting (≈ 10 GHz) is smaller than the separation between neighboring manifolds. It is well known that even for hydrogen the Hamiltonian is not separable when atomic diamagnetism is included.[2] Therefore, the approach suggested by Harmin cannot be applied here because it requires the hydrogen problem to be solved. In order to identify the experimentally observed signals, we have diagonalized the corresponding Hamiltonian H_d (Ref. 2) within a limited spherical basis set. Matrix elements of H_d were calculated within the semiclassical approximation including quantum defects.[16] Since the intermediate and final (Ryd-

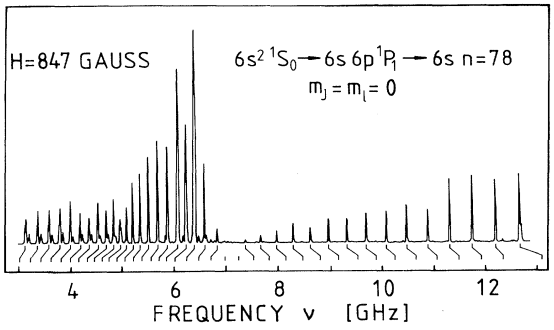

FIG. 5. Diamagnetic manifold ($n = 78$) of Ba. Intensities (vertical axis) are given in arbitrary units. The origin of the frequency scale corresponds to the $6s82s\ ^1S_0$ state in zero field [$E = 42016.776(2)\ \mathrm{cm}^{-1}$]. In the lower part, calculated line positions are indicated.

berg) states are pure singlets to a good approximation, $m_j = m_l$ and atomic paramagnetism can be neglected. In Fig. 5 we compare calculated line positions to the experimental spectrum. Although we have used the smallest possible basis set consisting of the states $80d$, $82s$, $78g$, 78 ($l = 6, 8, \ldots, 76$), $81d$, and $83s$, good agreement was obtained. Because of the truncated basis set, the calculated overall splitting is too large. A calculation of intensities as well as more accurate line positions would require a considerably enlarged basis set. The distribution of oscillator strengths within the diamagnetic manifold, shown in Fig. 5, is strikingly similar to those observed [cf. Fig. 3(b)] and calculated (cf. Fig. 4) for the $n = 78$ Stark manifold. The pronounced minimum in intensity (see Fig. 5) which is absent when we excite the $m_l = +1$ or $m_l = -1$ components, occurs for lines having the maximum $6s82s$ character. Analogous to the Stark spectra discussed above, at the minimum (cf. Fig. 5) s and d contributions to the transition moments destructively interfere.

In summary, we have excited Ba Rydberg states with principal quantum numbers up to $n = 290$. In addition we have studied barium Rydberg atoms with principal quantum numbers $70 \leq n \leq 160$ in external electric and magnetic fields. Main emphasis has been put on a comparison between interference effects observed for Stark as well as diamagnetic manifolds caused by the presence of a nonhydrogenic state. Furthermore, by studying Rydberg atoms with $n \gtrsim 100$ in external fields of 60 kG, our experiment offers the prospect of increasing the ratio of the diamagnetic force to the Coulomb force acting on the Rydberg electron by at least one order of magnitude.

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[a]Permanent address: Lund Institute of Technology, S-22100 Lund, Sweden.

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