LASER SPECTROSCOPY OF DOUBLY-EXCITED STATES IN BARIUM

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ABSTRACT

With pulsed and CW laser radiation we investigated doubly-excited, autoionising states of Ba. 4fnf series were excited from metastable $5d^2$ states using two pulsed dye lasers and 5dnf intermediate states. To explain the data on high-lying states knowledge of the wave functions of the intermediate states and their perturbers is prerequisite. In preparation of experiments on double circular states such as $4f^2$, 4f5g, $5g^2$ etc. we studied the 5d5g multiplet, excited with two pulsed lasers from 6s5d metastable states. Furthermore we present results of a study of 6pn$l$ states with large l, excited with two narrowband CW lasers. 6sn$l$ states were populated in a beam from metastable $5d^2$ states in the presence of an electric field. The atomic motion was used to reduce the field adiabatically to zero before applying the second excitation step (Stark switching). The results show that further excitation to 7dn$l$ and even Nfn$l$ states is feasible, giving possibilities to study the three-body Coulomb problem with CW laser resolution.

INTRODUCTION

The independent-electron model has been very successful in the description of the properties of atomic ground and singly-excited states. When two electrons are excited simultaneously outside their valence shell a more complex situation arises. Then the repulsive force between the electrons may be equal to the attractive force between each electron and the positively-charged 2^+-core and the motion of the electrons will show strong angular and radial correlations. Under these circumstances two-electron wave functions can no longer be expressed as simple products of two one-electron functions, i.e. the description of a state in terms of independent-electron quantum numbers breaks down. The study of this three-body Coulomb problem gained much in importance after the experiment of Madden and Codling[1] on doubly-excited states of Helium, which resulted in numerous experimental and theoretical contributions to the subject.

In recent years much effort has been invested in laser spectroscopic studies of the three-body Coulomb problem. Unfortunately the doubly-excited states of the He atom itself are inaccessible for laser excitation. For this reason experimental efforts concentrate on doubly-excited, autoionising states of the alkaline-earth atoms Ca, Sr and Ba[2-7]. The two loosely bound valence electrons are promoted to high-lying states in multi-step laser excitation processes. A problem is the influence of the non-spherical potential of the extended 2^+-core, which removes the degeneracy in the core-penetrating orbits with angular momentum $l<4$. The interaction of the penetrating electron with the core obscures the effect of the electron-electron repulsive interaction $1/r_{12}$. This makes it hard to observe correlation effects, although effects of positional correlations could be identified in barium ndNp double Rydberg states recently[8]. The creation of a heliumlike three-body Coulomb system in alkaline-earth atoms requires the elimination of these core-effects, i.e. the excitation of high-l states. Camus et al.[5] excited $6p_{3/2}nl$ and $6d_{5/2}nl$ states of Ba with $l=6$ - (n-1) and n=11-15 as well as $7d_{5/2}nl$ states with $l=7$ - (n-1) and n =11 - 14 using multi-step pulsed laser excitation and a Stark switching technique. Roussel et al.[9] produced in crossed electric and magnetic fields circular 6p21*l* states with *l* =m=20 in Ba. Double circular states 4f5g with total angular momentum J=3 in Ba were populated in a three-step laser excitation scheme by Jones et al.[4]. They observed low autoionisation rates, which might be an indication of strong radial correlation effects.

In doubly-excited N*l* n*l*" states two classes may be distinguished. One class contains states with N<<n. The electrons move in orbits with different radii r_1 and r_2. These states are called "valley" (or planetary) states as the electrons move in the valley of the six-dimensional Coulomb potential[10]. The majority of experiments on doubly-excited states concerns these valley states, but in most cases correlation effects were obscured by core-penetration effects. The second class involves so-called "ridge" states with N~n. Here both electrons move in orbits with nearly equal radii on the saddle of this six-dimensional potential. Then strong radial correlation effects are expected[10], but no clear experimental evidence has been found thusfar in laser experiments.

Our research thusfar mainly concentrated on doubly-excited, valley states of Ba populated in pulsed and CW multi-step laser excitation processes from metastable states of the 6s5d and $5d^2$ configurations. We studied 5dn*l*, 6pn*l* and 4fnf autoionising series in detail[7,11-14] using pulsed lasers. In two-step excitations with CW lasers we have populated 6pn*l* states with high *l* -values (n=40, *l* up to 30) using a Stark-switching technique. We observed narrow autoionisation profiles (down to ~ 85 MHz), which will allow us to apply further excitation steps to e.g. 7dn*l* and Nfn*l* states with N ~ n. In this contribution we will discuss some of the results on 4fnf and 6pn*l* series as well as work in progress to excite double-circular states such as $4f^2$, $5g^2$ and 4f5g.

EXPERIMENTAL ARRANGEMENTS

In the experiments we use a well-collimated beam of Ba atoms. Metastable states are populated by running a DC-discharge between the barium-filled tantalum oven and a tungsten heating wire in front of it. About one percent of the atoms is transfered from the $6s^2$ ground state to the metastable 6s5d configuration near 10000 cm^{-1} and 10^{-2} percent to the $5d^2$ configuration around 24000 cm^{-1}. In the interaction region used in multi-step excitation processes the laser light perpendicularly intersects the atomic beam to eliminate Doppler effects to a large extent (in experiments with CW, frequency-stabilised lasers down to about 10 MHz) and Ba atoms are directly excited to high-lying states above the first ionisation limit. Electrons released from the subsequent autoionisation process are counted with an electron multiplier. Count rates are stored on a computer, which controls the frequency of the scanning laser used in the last excitation step as well. Using several grids and voltages we can select the energy of the detached electrons and so discriminate between the various competing autoionisation channels. Great care was taken to eliminate stray electric fields in this interaction zone. Various excitation schemes to populate doubly-excited states, all starting at metastable levels of the 6s5d- and $5d^2$- configuration, are shown in Fig.1.

In the case of Stark-switching experiments with multi-step CW laser excitation several interaction regions are used. In the first region 6sn*l* bound Rydberg states are laser-excited from the metastable $5d^2$ $^1G_4$ state in the presence of a weak DC electric field by selecting a component of the resulting angular momentum manifold. Because of their motion the excited, long-lived atoms leave the diverging field adiabatically. Downstream, in a field-free region, the inner 6s-electron is subsequently excited to a 6p-orbit using a second CW laser. The n*l* Rydberg-electron remains unaffected in this process (isolated-core-excitation). Further downstream the atoms that remained in the 6sn*l* state are field ionised and detected to lock the first laser to the transition.

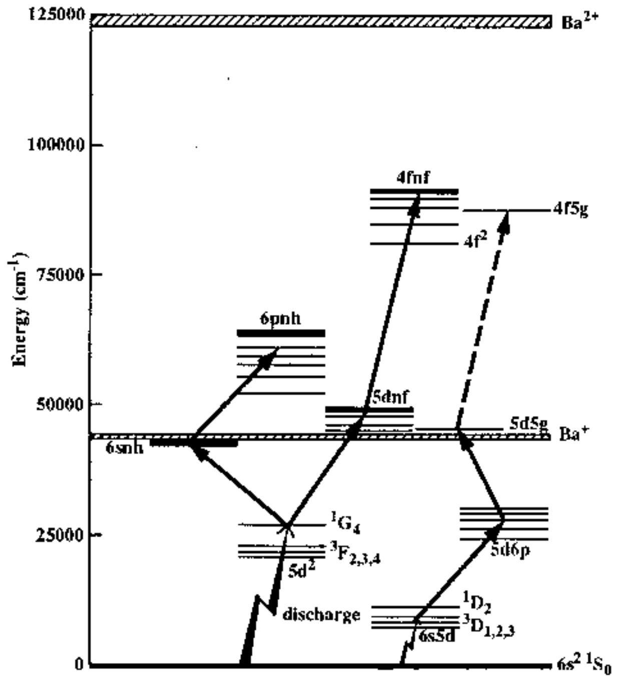

Fig.1. Excitation schemes used to populate high-lying, autoionising states in the barium atom.

PULSED LASER EXPERIMENTS ON DOUBLY-EXCITED SERIES

A. 4fnf series. Starting from metastable levels of the $5d^2$- configuration ($^3F_{3,4}$ and $^1G_4$ (see Fig.1)), we excited in a two-step process $4f_{5/2}nf$ J=2-6 states (n=9-46) and $4f_{7/2}nf$ J=3-6 states (n=19-21,25,40) with an energy of ~ 11eV above the ground state and ~ 5eV above the first ionisation limit [7]. In the first step 5dn'f states were excited with a Quanta Ray PDL3 pulsed dye laser with a bandwidth of 0.07 cm^{-1} in the wavelength range 380 - 470 nm. A frequency-doubled, Quanta Ray PDL2 laser with a bandwidth of 0.30 cm^{-1} was used for the next excitation step to 4fnf at ~230 nm. Both dye lasers were pumped with 355 nm light from a Nd:YAG laser (Quanta Ray GCR4). The linearly-polarised beams were overlapped in the interaction region.The principal quantum number of the excited 4fnf state is not necessarily identical to that of the intermediate 5dn'f state as a result of shake-up / -down processes.

5dnf J=4,5 series with extremely narrow autoionisation linewidths were earlier studied in detail by Bente and Hogervorst[11] using both pulsed and high-resolution CW lasers and many levels of J=2 and 3 series were identified in the present work. For J=5 the interaction between $5d_{3/2}n'f$ and $5d_{5/2}n'f$ series as well as interactions with the underlying continua were analysed in the framework of multi-channel quantum-defect theory (MQDT). Series perturbations and autoionisation linewidths were reproduced quite well. The $5d_{3/2}n'f$ J=4 series are, in addition, perturbed by levels of the $5d_{5/2}n''p$ series. This interaction was included in the analysis as well. Similar perturbations were also observed in the J=3 and 2 series but no attempt has yet been made to perform MQDT analyses. For the present experiments it is important to note that in the first excitation step with a pulsed laser to 5dn'f states in many cases it is not possible to resolve individual states, that the 5dn'f multiplet structure is not regular due to varying interactions with $5d_{5/2}np$ J-states and that their excitation probability is strongly influenced by these perturbations. This implies that for the interpretation of the data on 4fnf series it is extremely important to accurately know the energy positions of the 5dn'f J-states and the composition of their wave functions.

In Fig.2 spectra of $4f_{5/2}nf$ J=6,5,4 states, recorded in excitation from resolved $5d_{3/2}15f_{7/2}$J=5, $5d_{3/2}15f_{5/2}$J=4 and $5d_{3/2}15f_{7/2}$J=4 intermediates, are shown. In the excitation from J=5 a sharp resonance with laser linewidth was observed. It could be identified as a mixture of $4f_{5/2}16f_{7/2}$ and $4f_{7/2}13f_{5/2}$ J=6 states. This is a general feature of the excitation from J=5. The 4fnf states in Fig.2 are labeled according to a jj-coupling scheme $[n;j_1j_2J]$.

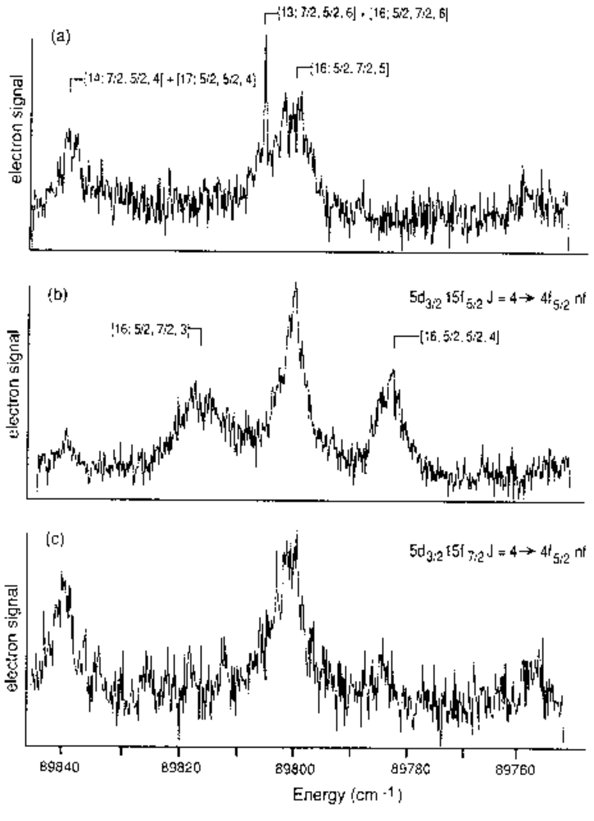

Fig.2. Spectra of $4f_{5/2}nf$ J=6,5,4 states in excitation from $5d_{3/2}15f$ J=5,4 states. 4fnf states are labeled $[n;j_1j_2J]$. Note the narrow J=6 resonance with dominant $4f_{7/2}nf$ character.

There are twelve $4f_{5/2}nf$ series (one J=6,0 and two J=5-1) converging to the $4f_{5/2}$-limit at 90239 cm^{-1} and fourteen $4f_{7/2}nf$ series (one J=7,0 and two J=6-1) converging to the $4f_{7/2}$-limit at 90518 cm^{-1}. The $4f_{7/2}n'f$ states interact with the $4f_{5/2}nf$ series. As the fine structure splitting of the 4f-ionic state is small (~225 cm^{-1}) many $4f_{5/2}nf$ states will be strongly mixed with $4f_{7/2}n'f$ states, which makes assignment difficult.

Furthermore interaction with 7pnf series converging to the $7p_{1/2}$-limit at 91424.95 cm^{-1} (four series, one J=4,2 and two J=3) and to the $7p_{3/2}$-limit at 92046.12 cm^{-1} (eight series, one J=5,1 and two J=4-2) may occur. The 4fnf series autoionise into several continua. For $4f_{5/2}$nf states 6d, 5d, 6p, 7s and 6s continua are available; for $4f_{7/2}$nf states above the $4f_{5/2}$-limit also the $4f_{5/2}$ continuum becomes available. Decay into the 6s or 7s continuum is unlikely because of the large change in angular momentum involved. In the experiment only electrons with an energy larger than 2 eV were detected, so decay into 6d, 7s and $4f_{5/2}$ channels could not be observed. As the different 4fnf J-states may all have different autoionisation branching ratios (e.g. they depend on admixture of 7pnf character) it was of limited value to compare relative intensities in the spectra.

Despite all these complications we succeeded in identifying and assigning 12 series. Helpful in the analyses was to graphically relate strong 4fnf resonances to series using the detailed knowledge on the intermediate 5dn'f states. As an example two Lu-Fano plots for J=5 and J=6 4fnf series are shown in Fig.3. Strong interactions show immediately in these plots. In the J=5 plot two perturbed $4f_{5/2}$nf series with reduced quantum defects of 0.1 and 0.8 appear. The series with defect 0.1 strongly interacts with the state $7p_{3/2}8f_{7/2}$ J=5 whereas the series with defect 0.8 interacts with $4f_{7/2}$nf states. The J=6 plot shows two series with quantum defects 0.54 and 0.68. The single $4f_{5/2}nf_{7/2}$ series interacts with states belonging to one of the two $4f_{7/2}$nf J=6 series.

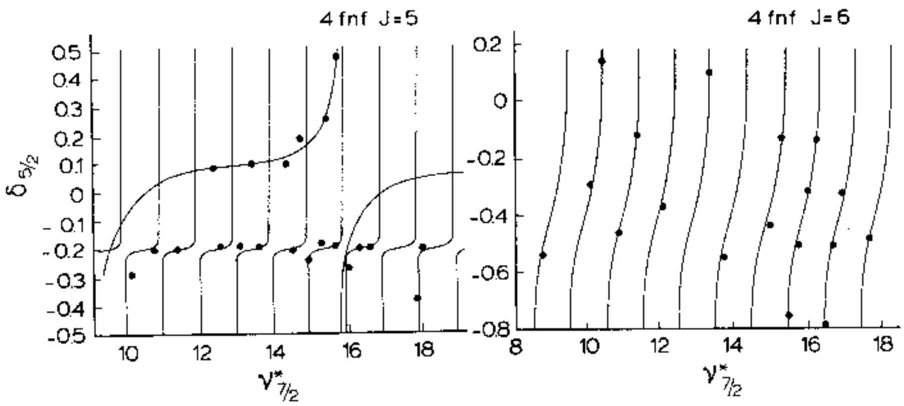

Fig.3. Lu-Fano plots of 4fnfJ=5 and 6 resonances. The J=5 plot (left) shows evidence of a strong interaction of the $4f_{5/2}$nf series with reduced quantum defect 0.1 with $7p_{3/2}8f_{7/2}$ J=5. The J=6 plot (right) shows a strong interaction of the $4f_{5/2}nf_{7/2}$ series with a $4f_{7/2}$nf J=6 series.

This and similar information on other series formed the basis for extensive MQDT analyses for each J - value observed. We also performed a Slater - Condon analysis of "unperturbed" energy values for a fictitious 4fnf multiplet at n=15 derived from the eigen quantum defects obtained from the MQDT analyses. When the spin-orbit interaction parameter ζ_{4f} is set equal to the ionic value (64.2 cm^{-1}) and ζ_{nf} to zero it turns out that only four electrostatic integrals (F^0, F^2, G^0 and G^2) are needed to reproduce the energy levels and their ordering in the multiplet. To eliminate the modulo

1 ambiguity in the quantum defects the energies must be related to the lowest members in the series, i.e. to the $4f^2$ configuration. These levels are still unknown, although in a preliminary experiment we tried to localise them. However, the nf-series in Ba^+ have quantum defects of ~0.9 and in elements with partially filled 4f-shell values of ~ 1.0 have been found. The nf-electron in Ba^+ moves in the combined Coulomb and centrifugal potential, showing two minima. The nf-wave functions are localised in both minima. This results in large phase-shifts of the radial wave functions and so in large quantum defects for nf-states. For this reason we tried combinations of quantum defects between 0.0 and 1.5 in the Slater-Condon fit of the 4fnf multiplet. The outcome of the fit is a widely spread multiplet with quantum defects between 0.0 (J=0) and 1.34 (J=4). The quantum defect interval for the 4fnf configuration for J=3-6 together with the corresponding intervals for the 6snf[15], the 5dnf[11] and 6pnf[13] configuration is given in Table I. Included as well is the interval (for J=3-6) for the 7pnf configuration that was deduced from the MQDT analyses of the 4fnf series[7].

Table I Quantum defects of Nlnf series of Ba

Configuration	Quantum defects	J-values
6snf	0.14 - 0.18	all
5dnf	0.05 - 0.14	1,4,5
6pnf	0.08 - 0.34	1 - 5
7pnf	0.20 - 0.60	3 - 6
4fnf	0.54 - 1.34	3 - 6

In Nl nf series of Ba with $l \leq 2$ the inner Nl -electron partly screens the 2^+ charge of the core for the nf-electron. As a consequence these series have much smaller quantum defects (see Table I) than the nf-series in Ba^+ (~0.9). The screening of the nf-electron by the 4f-electron is less effective and 4fnf states will be extremely sensitive to electron correlation effects. We ascribe the large increase in quantum defect values when the inner electron is excited to a 4f-orbit (see Table I) to these correlation effects. This was confirmed in ab initio calculations on the 4fnf J=5,6 series by Luc-Koenig and Aymar[16]. In general they find good agreement between calculated and measured level energie; the only difference is that a J=5 series tentatively assigned 7pnf by De Graaff et al.[7] probably is 4fnf. These calculations show that in the 4fnf configuration large quantum defects occur due to strong centrifugal barrier and two-electron correlation effects.

B. The 5d5g-configuration. In preparation of experiments on double circular states in Ba such as $4f^2$, 4f5g and $5g^2$ we have investigated the 5d5g multiplet just above the first ionization limit in detail. The bound 5d4f multiplet is known from earlier work on 6snh Rydberg series[17]. By exciting the 5d-electron of these multiplets in a one- or two-photon process circular states may then be reached.

We have excited the different states from the 5d5g multiplet in a two-step process using pulsed lasers, following the same procedure as in earlier experiments on 5dns, 5dnd and 5dng Rydberg series[12]. We use the metastable $6s5d\,^1D_2$, $^3D_{1,2,3}$ levels near 10.000 cm^{-1} to excite the various J-levels of the 5d6p-configuration around 25000 cm^{-1} (see Fig.1). In the next excitation step we populate the different 5d5g levels near 43300 cm^{-1} through an admixture of 5d4f character in the wave function of the 5d6p configuration. An example is shown in Fig.4, where a spectrum recorded in the excitation $6s5d\,^3D_2$ --> $5d6p\,^3D_2$ --> $5d_{5/2}5g$ is reproduced. The peaks are identified in a [jl]K-coupling scheme. The K= 3/2 and less so the K = 5/2 resonance show Fano-profiles, indicating interaction with, in this case, nearby members of the 5d8d

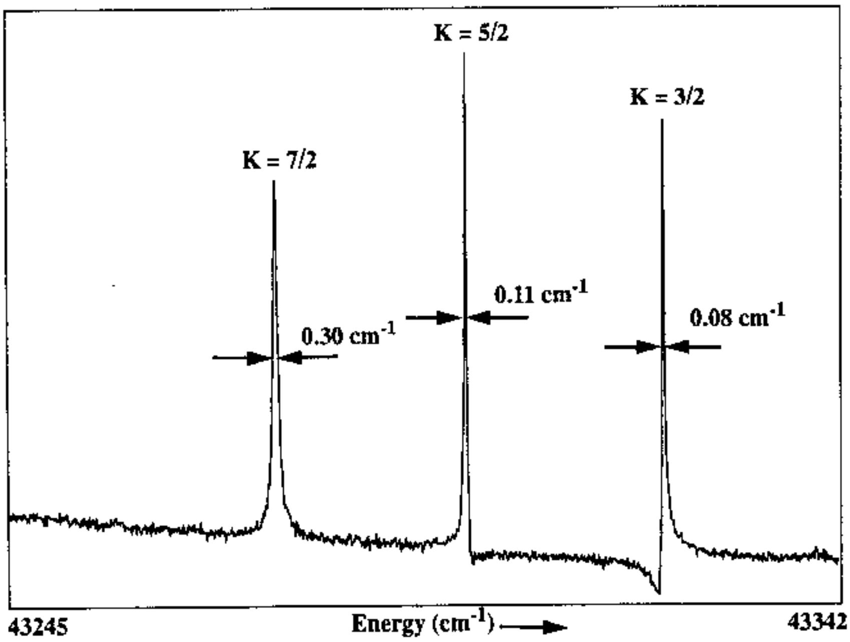

Fig.4. Excitation spectrum 6s5d $^3D_2$ --> 5d6p $^3D_2$ --> $5d_{5/2}5g$ using two pulsed dye lasers

multiplet. The rising "background" towards lower energy is the wing of the $5d_{3/2}8d$ J=2 resonance, interacting with the $5d_{5/2}5g$ K=3/2 peak. The K=3/2, 5/2 levels can only autoionise into 6sεd continua, whereas the higher K-levels only decay to 6sεg. Decay of 5d5g in 6sεg is much faster than in 6sεd as is obvious from autoionisation linewidths of the resonances shown e.g. in Fig.4 ; the linewidths of the K=3/2 and 5/2 peaks being laser linewidth.

We have observed ten (with J=2-5 and K=3/2-9/2) out of twenty possible 5d5g levels (eight $5d_{3/2}ng$ and twelve $5d_{5/2}ng$). With two additional levels (observed by Camus et al.[18]) a tentative Slater-Condon analysis of the 5d5g configuration could be performed. The experimental level energies were reproduced (on average) to far within 1 cm^{-1}, which indicates that the interaction with 5d8d levels, although apparant in some of the line profiles, does not affect the positions isignificantly. Fixing the spin-orbit constant of the 5d-electron at the ionic value ζ_{5d}=320.390 cm^{-1} and taking ζ_{5g} zero the electrostatic parameters deduced are: E_{AV} =42.969.85 (0.27) cm^{-1}, F^2=155.8 (2.1) cm^{-1}, F^4=10.9 (6.0) cm^{-1} and G^2= 8.0 (6.8)cm^{-1}. Varying ζ_{5g} did not change the outcome as in the fit also a value of zero within the error limits resulted.

CW LASER EXPERIMENTS ON DOUBLY-EXCITED SERIES

In two-step processes with frequency-stabilised ring dye lasers (Spectra Physics 380D), pumped by Ar^+-lasers, we excited series converging to the $6p_{3/2}$-ionic limit. Our goal was to identify 6pn*l* series with sufficiently long autoionisation lifetimes, so that they can be used for further excitation to 7dn*l* and, eventually, Nfn*l*. Starting from the metastable $5d^2\,{}^1G_4$ level we excited in a first experiment (see Fig.1) bound 6snh J=5 Rydberg states with a Rhodamine 6G dye laser (~600 mWatt). Here we use the small admixture of 6s5g character in the wavefunction of the metastable level. The laser is actively stabilised on these strong 6snh signals before applying the next excitation step to $6p_{3/2}$nh states using a Stilbene 3 blue dye laser. As an example a n=40 spectrum as a function of the frequency of the second laser (linewidth 1 MHz) is shown in Fig.5. Three components of the $6p_{3/2}40h$ multiplet with K=13/2 (strong), 11/2 and 9/2 (weak) were recorded (excitation from $6s_{1/2}40h_{11/2}$ J=5). Autoionisation linewidths could be determined accurately and turn out to be different for the various components. Although these 6pnh resonances are quite narrow, they appear to be too large to be used for further excitation with another CW dye laser. As states with $l < 5$ autoionise even faster we focussed attention on possibilities to excite 6pn*l* states with $l > 5$ in a second experiment.

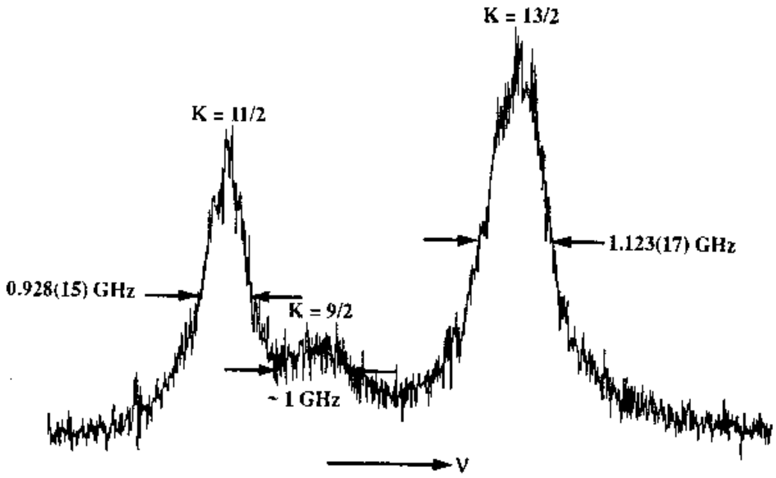

Fig.5. Spectrum of $6p_{3/2}40h$ excited with CW lasers in a two-step process from $5d^2\,{}^1G_4$ via the $6s_{1/2}40h_{11/2}$J=5 bound Rydberg state. Resonances are assigned in [j*l*]K coupling.

We changed our CW laser setup to implement the technique of Stark-switching. This technique is now commonly used in pulsed laser experiments to produce high-*l* angular momentum states (see e.g. ref. 5). In the presence of an electric field the linear Stark effect is used to excite a state with high value of the appropriate parabolic quantum number k. This field must decay adiabatically to zero to preserve in a field-free region the *l*-character corresponding to this k-state (see Fig.6). In pulsed laser experiments this is achieved by switching off the electric field sufficiently slow.

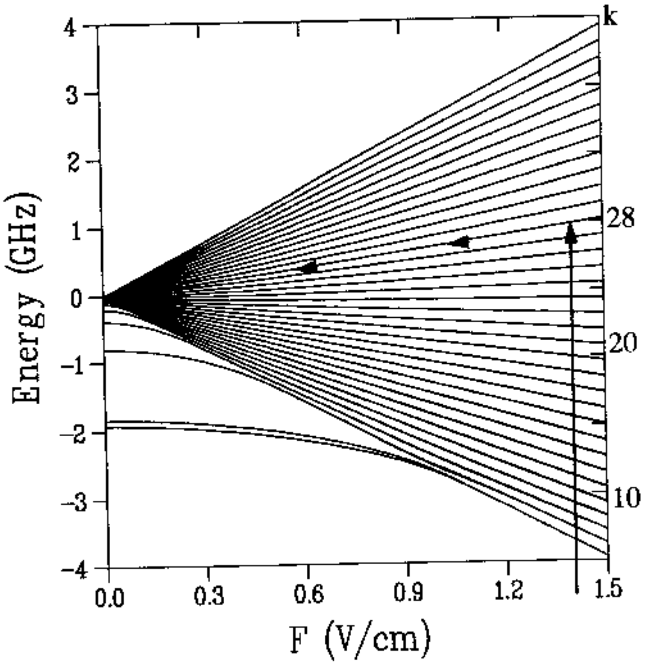

Fig.6. Calculated 6snk manifold as a function of electric field strength F. The arrows indicate the Stark-switching route used in experiments (see Fig.7).

In our experiments we use the thermal velocity of the Ba atoms (500 m/s). Ba atoms are excited to 6snk states in between a set of capacitor plates. The electric field strength is chosen such that individual k-components are excited. E.g. for n=40 a field of 1.5 V/cm is sufficient. Over a distance of about 7 mm (~14 μs) the atoms move out of the diverging electric field and enter a well-shielded field-free region for excitation to $6p_{3/2}nl$ states. The field gradient is such that the transition to the field-free region is adiabatic. In Fig.7 examples of excitations of $6p_{3/2}40l$ for different l are given. In the lower part the 6snk manifold recorded in an electric field of 1.4 V/cm is shown. The first laser was locked to k=10, 20 or 28 while the second, blue laser was tuned over the $6s_{1/2}$ --> $6p_{3/2}$ transition. In the upper part three spectra, identified with a k-vakue, are shown Important feature is that autoionisation linewidths decrease with increasing l -value to about 85 MHz at l =27 (-> k=28). This value is sufficiently small to use this state for further excitation, as planned in the near future, but not as low as expected from the decrease in overlap between the wave function of the $40l$ electron with the core. For high l -values autoionisation of $6p_{3/2}40l$ will be strongly inhibited and radiative decay to $5d40l$ will be the dominant process. Eventually $5d40l$ autoionises at a low rate[11]. This implies a minimum observable width of 20 MHz in our experiment, corresponding to the radiative decay time of $6p_{3/2}40l$. The observation of larger widths is probably due to the presence of small stray-electric fields of a few mV/cm in the field-free region, that induce mixing of states with different l -values. From the high sensitivity of highly-excited states for electric fields it can be concluded that in experiments where such states, in particular with $l \geq 3$, are involved extreme care has to be taken to eliminate stray fields. In low-resolution pulsed laser experiments such fields might easily induce the excitation of many l states in the manifold.

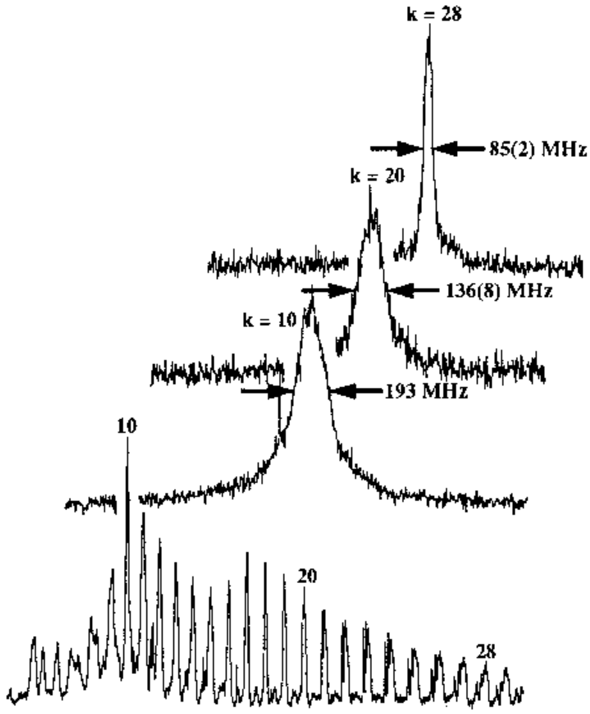

Fig.7. Two-step excitation of $6p_{3/2}40l$ from $5d^2\ ^1G_4$ via 6snk. These latter states were excited in a field of 1.4 V/cm (see Fig.6).

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