

## The $5dnp$ and $5dnf$ $J = 1$ autoionizing series of Ba in one-photon laser excitation from the $6s^2\ ^1S_0$ ground state

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**Abstract.** The  $5dnp$ ,  $nf$   $J = 1$  series of barium converging to both the  $5d_{3/2}$  and  $5d_{5/2}$  limits were studied in a heat-pipe system in a one-step pulsed UV laser excitation from the atomic ground state. Strong interseries and continuum interactions were observed and experimental evidence of interaction with  $6p7s$   $J = 1$  levels was found. A comparison of the observed spectra with spectra calculated using an eigenchannel  $R$ -matrix multichannel quantum-defect method is presented. The agreement is satisfactory in view of the complexity of the calculations.

### 1. Introduction

Interest in laser spectroscopy of atoms with two valence electrons such as barium is more and more focusing on a study of high-lying doubly excited states or double Rydberg states (Camus *et al* 1989, Eichmann *et al* 1990, Jones 1990). These states are populated using multistep laser excitations, where each step or most of the steps are resonant with atomic transitions. Often bound Rydberg states or low-lying autoionizing states play an important role as intermediate levels in any excitation scheme. To facilitate the explanation of the observations on high-lying doubly-excited states it is of importance to know in detail the character of these intermediate states.

As part of a program to study in detail low-lying autoionizing series of the type  $5dnl$  and  $6pnl$  of barium results on the odd-parity  $5dnp$  and  $5dnf$   $J = 1$  series converging to the  $5d_{3/2}$  and  $5d_{5/2}$  ionization limits and excited directly from the  $6s^2\ ^1S_0$  ground state will be reported. Recently results of an investigation of  $5dnf$   $J = 4$  and 5 series with cw laser resolution were published (Bente and Hogervorst 1989a). Even-parity  $5dnl$  series were, for example, extensively studied by Camus *et al* (1983) and Bente and Hogervorst (1989b). For recent work on  $6pnl$  series reference is made to de Graaff *et al* (1990), Carré *et al* (1990) and Lange *et al* (1991). Also recently the full manifold of  $6pnf$   $J = 1-5$  series was studied, enabling an analysis of the multiplet fine structure (Abutaleb *et al* 1991). Many of these states are now being used as intermediate levels for the excitation of even higher-lying doubly-excited configurations (see e.g. Jones 1990). The use of the  $5dnf$  configuration is especially promising as it can be reached easily directly from the ground state or from metastable states of the  $5d^2$  configuration at approximately 3 eV above the ground state (Vassen *et al* 1989).

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The  $5dnp$  and  $5dnf$   $J=1$  series of Ba were studied earlier in the well known classical absorption experiments of Garton and Tomkins (1969) using the  $6s^2\ ^1S_0$  atomic ground state. Although precise values of the different series converging to the  $5d_{3/2}$  and  $5d_{5/2}$  ionization limit were given, experimental resolution was not sufficient to observe in full detail, e.g., line profiles of these interacting autoionizing series. Using synchrotron radiation Griesmann (see Aymar 1990) studied the same series, again in low spectral resolution. Recently Gounand *et al* (1991) studied the  $5dnp$  and  $5dnf$   $J=1$  series in a pulsed laser experiment starting from the  $5d^2\ ^1S_0$  level at  $25\,873.9\text{ cm}^{-1}$ . This state is efficiently populated in the decay from, e.g., the  $6s10p\ ^1P_1$  bound Rydberg level, enabling its use as an intermediate level for the excitation of  $5dnp$  and  $5dnf$  series. They used the  $R$ -matrix multichannel quantum-defect (MQDT) calculations of Aymar (1990) to compare with the experimental results. The difference between the experiment of Gounand *et al* (1991) on the one hand and the uv absorption experiment of Garton and Tomkins (1969) and the pulsed laser experiment reported here on the other, lies in the properties of the initial state used for the excitation. The variational  $R$ -matrix calculations of Aymar (1990) show that the  $5d^2\ ^1S_0$  state has about 72.5%  $d^2$ , 20%  $p^2$  and 7.5%  $s^2$  character, whereas the  $6s^2\ ^1S_0$  ground state has a composition of 92%  $s^2$ , 6.7%  $p^2$  and 1.3%  $d^2$ . In both cases it is the  $d^2$  character of the states that is excited, but the spectra observed, in particular the intensity ratios, are quite different reflecting differences in the wavefunctions of the initial state. Furthermore, from the dominant  $s^2$  part of the ground state direct excitation of the  $6s\epsilon p$  continuum occurs, giving rise to pronounced interference effects (Beutler-Fano profiles). The continuum is only weakly excited in spectra taken from the  $5d^2\ ^1S_0$  state because of the small admixture of  $s^2$  character. This implies that ground-state measurements provide a more sensitive test for oscillator strength distributions as well as for interactions of  $5dnp$  and  $5dnf$  states with the  $6s\epsilon p$  continuum and as such are complementary to the experiments starting from  $5d^2\ ^1S_0$ .

In the following the experimental set-up and results of the one-photon excitation experiments on  $5dnp$  and  $5dnf$   $J=1$  autoionizing series will be presented first (section 2). In section 3 the data will be discussed and compared with theoretical spectra obtained by Aymar (1990) using eigenchannel  $R$ -matrix MQDT calculations. Finally in section 4 some conclusions will be drawn.

## 2. Experiments and results

In the present experiment barium atoms were excited with a single laser photon from the  $6s^2\ ^1S_0$  ground state to  $5dnp$  and  $5dnf$   $J=1$  autoionizing states in the energy range  $45\,860\text{--}47\,710\text{ cm}^{-1}$ . The  $5d_{3/2}np$  ( $n > 14$ ) and  $5d_{3/2}nf$  ( $n > 11$ ) series converging to the  $5d_{3/2}$  limit at  $46\,908.75\text{ cm}^{-1}$  as well as the  $5d_{5/2}np$ ,  $nf$  series converging to the  $5d_{5/2}$  limit at  $47\,709.73\text{ cm}^{-1}$  were studied. Pulsed laser light with a bandwidth of  $0.5\text{ cm}^{-1}$  in the wavelength range  $205\text{--}220\text{ nm}$  was generated by frequency doubling a Quanta Ray PDL2 dye laser operating on Stilbene 3 with a beta-barium-borate (BBO) crystal. The dye laser was pumped with the third harmonic of a Quanta Ray DCR3 Nd-YAG laser at  $355\text{ nm}$ . The  $5dnp$ ,  $nf$  series were excited with a uv laser of power of the order of  $0.01\text{ mJ/pulse}$ . Care was taken to avoid saturation effects in the excitation process. The wavelengths of the visible output of the tunable dye laser were measured in a home-built echelle grating wavelength meter (accuracy  $0.2\text{ cm}^{-1}$ ). Laser scans were calibrated using a Fabry-Perot interferometer with a free spectral range of  $1\text{ cm}^{-1}$ .

The barium atoms were excited in a heat-pipe system equipped with quartz windows for transmission of the (unfocused) UV laser light. The heat-pipe was heated to a temperature of about 1000 K to create a suitable barium density. The resulting Doppler broadening ( $<0.1 \text{ cm}^{-1}$ ) was small compared with the laser linewidth. Argon buffer gas (0.1 mbar) was added to prevent condensation of barium vapour onto the windows as well as to control the length of the interaction zone inside the hot part of the heat-pipe. To ensure field-free excitation a thermionic ring diode configuration as described by Beigang *et al* (1984) was used. It consists of eight cathode wires arranged radially symmetric parallel to the axis of the heat-pipe. The electric field produced by these wires is exactly zero in the centre of this ring configuration. This arrangement also increases the sensitivity of detection of ions produced by the laser excitation compared with a heat-pipe with a single cathode wire. For each laser pulse the voltage change over a load resistance of  $47 \text{ k}\Omega$  between cathode wires and heat-pipe wall was measured. A negative bias voltage of about 1 V was maintained over the thermionic diode, which operated in the space-charge limited current mode. The laser-induced ion signal was gated and integrated using a boxcar integrator; averaging over six laser shots was performed using a VME computer.

Examples of spectra recorded are shown in figures 1 and 2. In figure 1 the spectrum of the  $5d_{5/2}np, nf$   $J=1$  autoionizing series in between the  $5d_{3/2}$  and  $5d_{5/2}$  ionization

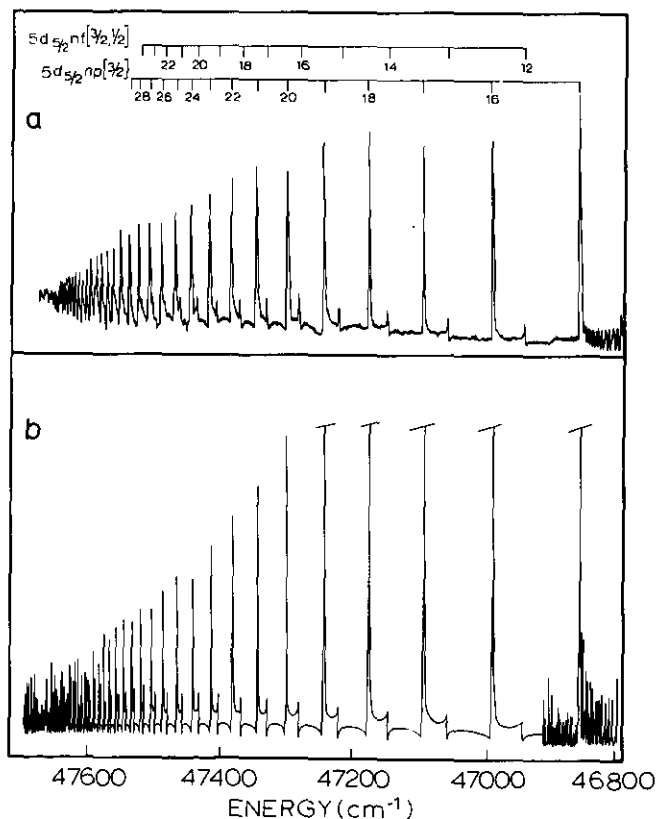


Figure 1. Recorded spectrum of the  $5d_{5/2}np, nf$   $J=1$  series of barium above the  $5d_{3/2}$  ionization limit at  $46908.75 \text{ cm}^{-1}$  (a) as well as the spectrum calculated with the eigen-channel  $R$ -matrix MQDT method (b).

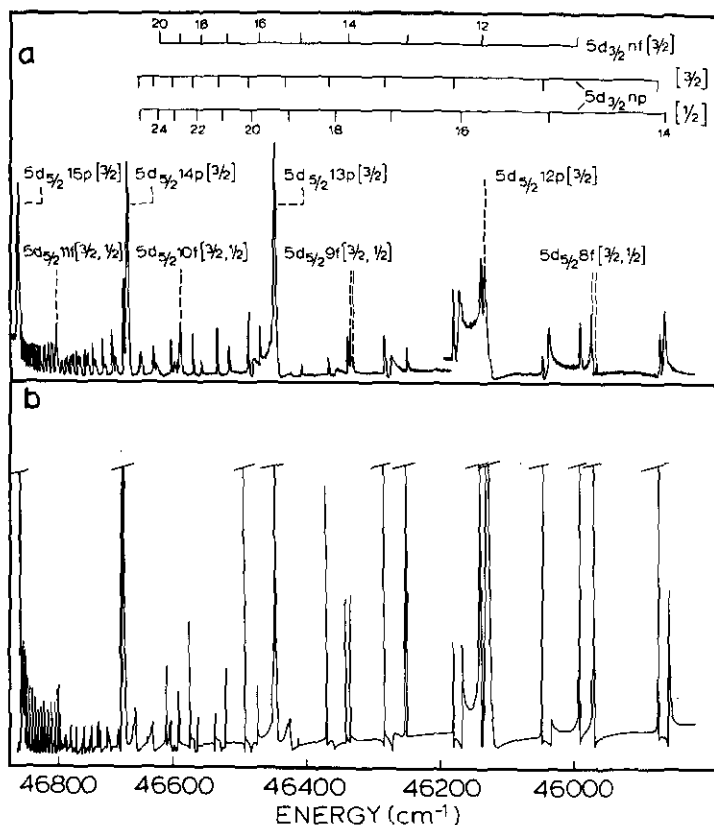


Figure 2.  $5dnp$ ,  $nf J=1$  series of barium below the  $5d_{3/2}$  ionization limit. (a) Recorded spectrum and (b) calculated spectrum. In the experimental spectrum the various  $5d_{3/2}$  and  $5d_{5/2}$  series are identified. A small part of the spectrum close to the  $5d_{3/2}$  limit is included in figure 1.

limits is given, whereas in figure 2 the spectrum below the  $5d_{3/2}$  limit is reproduced. These spectra differ in several aspects from the spectra reported by Gounand *et al* (1991) in their figures 2 and 3. They excited the  $5dnp$ ,  $nf J=1$  series from the  $5d^2 \ ^1S_0$  metastable state, resulting in strongest signals for  $5dnf$  states, whereas in the present experiment the  $5dnp$  states were dominantly excited from the  $6s^2 \ ^1S_0$  ground state. This is remarkable as in both cases the  $d^2$  character of the levels is excited. Furthermore, in excitation from the metastable state with its small admixture of  $s^2$  character (Aymar 1990) direct excitation of the  $6s$  continuum is weak, whereas this continuum is strongly excited from the ground state. This resulted in pronounced Beutler-Fano interference profiles in the present experiment as shown in figures 1 and 2. Energy positions of levels observed in both experiments are in agreement within the experimental errors.

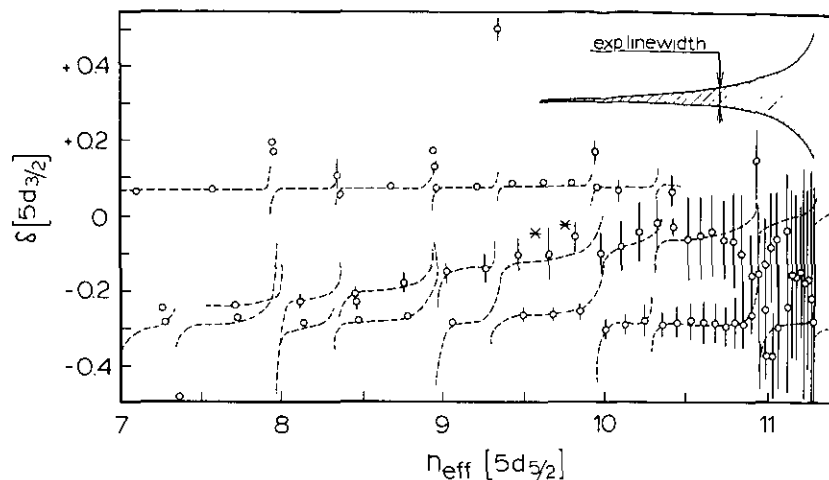
### 3. Discussion and analysis

In the  $5dnp$ ,  $nf J=1$  configuration of barium, six series may be excited: three series converging to the  $5d_{3/2}$  limit, in  $jK$  coupling  $5d_{3/2} np [\frac{3}{2}]$ ,  $5d_{3/2} np [\frac{3}{2}]$  and  $5d_{3/2} nf [\frac{3}{2}]$ , and three series to the  $5d_{5/2}$  limit, namely  $5d_{5/2} np [\frac{3}{2}]$ ,  $5d_{5/2} nf [\frac{1}{2}]$  and  $5d_{5/2} nf [\frac{3}{2}]$ . The

series converging to the  $5d_{3/2}$  limit are perturbed by members of the series converging to the  $5d_{5/2}$  limit and will autoionize into the two  $6s_{1/2}\epsilon p$  continua. For the autoionization of the states above the  $5d_{3/2}$  limit, five continua (two  $6s_{1/2}\epsilon p$ , two  $5d_{3/2}\epsilon p$  and one  $5d_{3/2}\epsilon f$ ) are available which could not be distinguished in this experiment. The  $6s_{1/2}\epsilon p$  continua were also excited directly from the 92%  $s^2$  character of the ground state as were  $5d_{3/2}$  continua, although much weaker, from its 1.3%  $d^2$  character. Strong interference effects between 'bound' and free channels will occur as was observed in the experimental spectra. Finally the lowest members of the  $6pns$   $J=1$  series, the  $6p_{1/2}7s$  and  $6p_{3/2}7s$  levels, might interact with the series studied. These levels are expected to lie below or close to the  $5d_{3/2}$  ionization limit, but no particular structure was observed thus far as they are expected to be completely smeared out over numerous resonances (Aymar 1990). The recent work of Lange *et al* (1991) on the  $6pns$   $J=1$  series does not involve the position of these levels. In the present analysis some evidence will be given that the  $6p7s$   $J=1$  levels do influence the  $5dnp$ ,  $nf$   $J=1$  series.

Experimentally above the  $5d_{3/2}$  limit only two distinct series could be observed (see figure 1). The strongest, slightly asymmetric lines belong to the  $5d_{5/2}np[\frac{3}{2}]$  series, whose lower members ( $n=15-12$ ) show up as equally strong features in the spectrum below the  $5d_{3/2}$  limit (figure 2). Their linewidths converge to the laser linewidth of  $0.5\text{ cm}^{-1}$  for high  $n$ . The weaker Beutler-Fano type profiles in figure 1 belong to the unresolved  $5d_{5/2}nf[\frac{1}{2}]$  and  $[\frac{3}{2}]$  series. The  $n=11-8$  lines of these series were also observed below the  $5d_{3/2}$  limit, as shown in figure 2, and the two components could be resolved for  $n=8$  and 9. Below the  $5d_{3/2}$  limit lines of the three  $J=1$  series converging to this limit were excited. The  $5d_{3/2}nf[\frac{3}{2}]$  series was observed from  $n=11-23$  with varying excitation strength but with a nearly constant linewidth, in agreement with the measurements of Gounand *et al* (1991). For higher  $n$  values the lines of this series could no longer be resolved from  $5d_{3/2}np[\frac{1}{2}]$  lines. This leads to an explanation of the difference of up to  $2\text{ cm}^{-1}$  in the measurements of Gounand *et al* (1991) and Garton and Tomkins (1969) for the energies of the  $5d_{3/2}nf[\frac{3}{2}]$  series for  $n>24$ . Garton and Tomkins obviously observed for  $n>24$  the  $5d_{3/2}np[\frac{1}{2}]$  series, so the level energies reported by Gounand *et al* for  $5d_{3/2}nf[\frac{3}{2}]$  are correct. The  $5d_{3/2}np[\frac{1}{2}]$  series was excited starting from  $n=14$  and also shows strong variations in excitation strength as well as unexpected and irregular changes in line profiles. Remarkable in this series is the increasing scaled linewidth, as calculated from fits to Fano profiles, with growing  $n$ . This is in agreement, both qualitatively and quantitatively when a comparison is possible, with the measurements of Gounand *et al* (1991) reported in their table 1. Similar observations were made for the  $5d_{3/2}np[\frac{3}{2}]$  series, which was also recorded starting at  $n=14$ .

The data for these 'bound'  $5d_{3/2}$  series are graphically represented in the Lu-Fano plot of figure 3. This quantum defect plot is to a large extent identical to the plot given by Gounand *et al* (1991). However, in the plot of figure 3 the experimental linewidths are included, represented by the vertical bars. For comparison the laser linewidth as a function of  $n$  is also given in figure 3. The remarkable features of this Lu-Fano plot are the strong energy dependence of the quantum defect of the  $5d_{3/2}np[\frac{1}{2}]$  series, the increase in linewidth with increasing  $n$  value in this series and maybe a similar increase in linewidth in the  $5d_{3/2}nf[\frac{3}{2}]$  series, although this is less clear as data for high  $n$  are lacking. These characteristics are probably due to interaction of these series with the  $6p7s$  levels as mentioned earlier. Here it is of interest to point out an analogy with the bound  $6snp\ ^1P_1$  series perturbed by the  $5d8p\ ^1P_1$  level (Aymar 1990). This level lies close to the  $6s_{1/2}$  limit and is smeared out over a large number of  $6snp$  states. This



**Figure 3.** Lu-Fano plot of quantum defects  $\delta(5d_{3/2})$  against the effective principal quantum number with respect to the  $5d_{5/2}$  limit,  $n_{\text{eff}}(5d_{5/2})$ , below the  $5d_{3/2}$  limit. Vertical lines through the experimental points indicate the linewidth of the resonance. The linewidths marked with an asterisk were determined from a fit of the resonances to a Beutler-Fano profile. For a comparison the experimental linewidth as a function of  $n_{\text{eff}}$  is included in this plot. The broken curves through the experimental points are to guide the eye.

results in a strong energy dependence of the quantum defect of the bound series as well as a growing oscillator strength with increasing  $n$  value in the ground-state transition  $6s^2\ ^1S_0 \rightarrow 6snp\ ^1P_1$  for  $n > 24$  (Connerade *et al* 1988). In Aymar (1990) it is shown in her figure 5 that the photoionization cross section close to and above the  $6s_{1/2}$  limit is clearly dominated by the presence of this  $5d8p\ ^1P_1$  level. A similar feature, although less pronounced, is observed in the present experiment just below the  $5d_{3/2}$  threshold, as shown in figure 1, but care has to be taken here as high  $n$  states are extremely sensitive, e.g., to very weak stray electric fields.

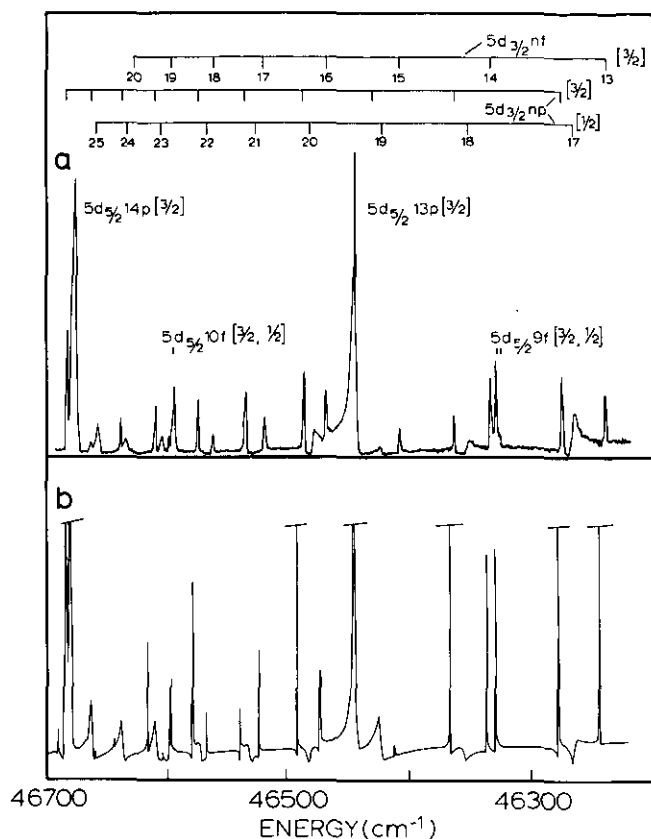
No effort was undertaken to fit all experimental data available using the phase-shifted reaction-matrix formalism of multichannel quantum-defect theory (MQDT) as was done, for example, in a previous paper on  $5dnf\ J = 4, 5$  series (Bente and Hogervorst 1989a). Although it was not too difficult to fit the spectra observed for the  $5d_{5/2}np$ ,  $nf\ J = 1$  series in between the  $5d_{3/2}$  and  $5d_{5/2}$  limit in a (simplified) five- or six-channel MQDT model (three series interacting with underlying continua) these results are of limited value. Below the  $5d_{3/2}$  limit the analysis is much more complicated and at least a ten-channel fit (when including  $6pns$ , but not  $6pnd$  channels) involving many parameters has to be performed. For this reason it was decided to confront the present barium data collected in excitation from the  $6s^2\ ^1S_0$  ground state with eigenchannel  $R$ -matrix MQDT calculations of Aymar (1990), as was also done by Gounand *et al* (1991) for their data recorded in excitation from the  $5d^2\ ^1S_0$  intermediate state. The calculational procedure is described in full detail in the original paper (Aymar 1990), whereas a summary is presented in the paper of Gounand *et al* (1991), which will not be repeated here. It suffices to mention that in the present calculations ten channels were involved, including the  $6pns$  channels. The differences in the calculated spectra in the excitations from  $5d^2\ ^1S_0$  and  $6s^2\ ^1S_0$  are due to the differences in dipole matrix elements  $D_a$  connecting these initial states to the  $R$ -matrix  $\Psi_a\ ^1P_1$  eigenstates. The  $D_a$  elements are strongly energy dependent and some typical values calculated at the  $5d_{3/2}$

threshold are given in table 1. Although the independent-particle model predicts the 5d-nf dipole matrix elements to be larger than those for 5d-np it appears that  $D_3(6s^2) \approx \frac{1}{4}D_4(6s^2)$ ; this probably results from cancellation effects related to the composition of the ground-state wavefunction. This explains the dominance of 5dnp peaks in photoionization from the ground state. The observation of strong 5dnf peaks in the excitation from the  $5d^2\ ^1S_0$  state is also explained since  $D_4(5d^2) > D_3(d^2)$  in table 1. In addition the  $D_2$  values associated with the  $6pns\ ^1P_1$  channel emphasize the large role of this channel near the  $5d_{3/2}$  threshold. In figures 1 and 2 the photoionization cross sections calculated in steps of  $0.3\text{ cm}^{-1}$  are included below the experimental spectra. These calculated spectra were not folded with the experimental linewidth. The overall agreement in the positions of the resonances is as good as in the case studied by Gounand *et al* (1991). Differences between experiment and calculations still occur in detail. However, it must be noted that the convergence of the variational calculations was not fully checked and that the shape of resonances is much more sensitive to the details of the two-electron basis functions introduced in the  $R$ -matrix calculations than the positions.

Experimental and calculated spectra above the  $5d_{3/2}$  limit as shown in figure 1 are in quite good agreement. Although the experimental linewidths of 5dnp resonances appear to be slightly larger than calculated values, this is compensated by larger peak heights in the calculations so that the total oscillator strength for 5dnp resonances is reproduced quite well. Below the  $5d_{3/2}$  limit (see figure 2) the calculations reproduce the complex structures near  $5d_{5/2}np$  resonances except for some calculated  $5d_{3/2}np[\frac{3}{2}]$  and  $5d_{3/2}nf[\frac{3}{2}]$  resonances ( $n=14-18, 20, 27, n=11-13$  respectively). They are too narrow and intense compared with the experimental lines, but again the differences in total oscillator strength are less pronounced. The calculations show that  $5d_{3/2}np[\frac{3}{2}]$  resonances are broader and weaker than  $5d_{3/2}np[\frac{1}{2}]$  and that only one  $5d_{3/2}19p$  resonance appears, in agreement with observations. However, some changes in intensity and line profile with  $n$  are not fully reproduced by the calculations. This is best seen in figure 4, which shows on an enlarged scale details of the experimental and calculated spectra in the energy range  $46\,200-46\,700\text{ cm}^{-1}$ . For example, in the  $5d_{3/2}np[\frac{1}{2}]$  series in the experimental spectrum when going from  $n=23$  to  $n=17$  the line shape changes from a Lorentzian to a Fano-type profile with a sign change in the Fano  $q$  parameter between  $n=19$  and  $18$ , which in the calculated spectrum occurs between  $n=18$  and  $17$ . In the  $5d_{3/2}np[\frac{3}{2}]$  series in the same energy range for several  $n$  values, calculated profiles differ from the experimental profiles. For example, for  $n=21-23$  the experimental lines have a near-Lorentzian shape, whereas the calculated lines show pronounced Fano-Beutler profiles. These differences indicate that the interactions of the bound  $5d_{3/2}np, nf\ J=1$  series with the underlying continua as well as with the  $6p7s$  levels slightly deviate from calculated interactions. Below the  $5d_{3/2}$  threshold ten channels are strongly interacting and strong interference effects will occur. So the shape and height of the resonances will be very sensitive to small variations in the parameters

Table 1. Dipole matrix elements calculated near the  $5d_{3/2}$  threshold.

$^1P\ \alpha$ channels	1 6snp	2 6pns	3 5dnp	4 5dnf
$D_\alpha(6s^2) = \langle 6s^2   D   \Psi_\alpha \rangle$	-1.4	-1.1	-2.8	0.70
$D_\alpha(5d^2) = \langle 5d^2   D   \Psi_\alpha \rangle$	-1.0	-1.2	-5.0	-10.0



**Figure 4.**  $5dnp, nf J = 1$  spectra in the energy range 46 200–46 700 cm<sup>-1</sup>. (a) Experimental spectrum and (b) calculated spectrum. This figure shows on an enlarged scale part of the spectra of figure 1.

which explains the difficulty in reproducing the experimental data in full detail. It is noteworthy to mention that an eight-channel *R*-matrix calculation (Aymar 1990), in which the  $6pns$  channels were not introduced explicitly, results in poorer agreement with experiment in the energy range studied in this work. This gives theoretical confirmation of the important influence of the  $6p7s$  levels on the line profiles.

It is interesting to note that recently eigenchannel *R*-matrix calculations in Ca (Farooqi *et al* 1991) have shown that the  $4p6s \ ^1P_1$  perturber is responsible for the observed fluctuations in the width of the  $3dnp \ ^1P_1$  resonances. In the same way for Sr (Aymar 1987) the interaction of  $5dnd$  and  $5pns$  channels with  $4dnp \ J = 1^0$  is responsible for anomalous variations in the intensity and width along the  $4dnp \ ^1P_1$  series.

#### 4. Concluding remarks

In the present experiment the  $5dnp, nf J = 1$  series in barium were studied in high resolution, using a single photon laser excitation process from the  $6s^2$  ground state. This investigation may be considered to be an improved version of the classical absorption experiment of Garton and Tomkins (1969). Also it is complementary to



recent work of Gounand *et al* (1991), who excited the same series from a metastable state of the  $5d^2$  configuration also using pulsed laser techniques. These two laser studies provided highly accurate information for a comparison with the eigenchannel  $R$ -matrix MQDT calculations of Aymar (1990). The overall agreement between experiment and calculations is satisfactory, certainly in view of the complexity of the calculations. In this paper emphasis was on the analysis of line profiles as it is obvious that differences between experiment and calculations will be most pronounced in the shape of resonances and not so much so in their positions. This indeed was the case in the comparison of experimental data with calculations. Evidence for interactions between  $5dnp$ ,  $nf$   $J=1$  series with  $6p7s$   $J=1$  levels could be deduced from the experimental data, more in particular from the strongly energy-dependent quantum defect of the  $5d_{3/2}np[\frac{1}{2}]$  series, the anomalous line broadening with increasing  $n$  within the same series and the behaviour of the Fano  $q$  parameter along the  $5d_{3/2}np[\frac{1}{2}, \frac{3}{2}]$  series, as well as from the  $R$ -matrix calculations. The present investigation confirms that this interaction may be a general feature in the heavy alkaline-earth atoms as indicated by the work of Aymar (1987) on Sr and of Farooqi *et al* (1991) on Ca.

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