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Autoionization Inhibited by Internal Interferences

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We have measured the linewidths of $[5d_{3/2}n_2d_{3/2}]_{J=0}$ autoionizing Rydberg states of barium with principal quantum numbers ranging between $n_2=14$ and $n_2=100$. Dramatic deviations of the autoionization rates from the normal n_2^{*-3} scaling law have been observed. Whereas the $[5d_{3/2}60d_{3/2}]_{J=0}$ level exhibits a linewidth Γ (FWHM) as low as 6 MHz, higher ($n_2 \approx 80$) members of this series are broadened up to 5 GHz. Autoionization is inhibited for principal quantum numbers $n_2 \approx 60$ because of configuration interaction between the $[5d_{3/2}n_2d_{3/2}]_{J=0}$ and $[5d_{5/2}n_3d_{5/2}]_{J=0}$ Rydberg series. This causes the state at $n_2=60$ to be metastable.

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High-energy autoionizing states have generated considerable interest recently as possible storage states for extreme-uv lasers.¹ By sharing energy between two excited electrons, it is possible for the state to store an energy greater than the atomic ionization limit. However, these states usually autoionize very rapidly ($\sim 10^{-14}$ sec) through the interelectron Coulomb interaction, so that interest has generally been directed towards metastable autoionizing states. Alkali-atom quartet states were the first observed examples of metastable autoionizing states²; autoionization is inhibited in these cases because the Coulomb interaction conserves the total spin S while all the possible final states of an ion plus a continuum electron are doublets. Similarly conservation of parity and total orbital angular momentum³ can also inhibit autoionization, although most of these selection rules begin to fail in heavy atoms because of spin-orbit coupling.

A variety of methods have been proposed for the use of external fields to create metastable autoionizing states. Strong laser fields⁴ have been predicted to produce metastable states by canceling out the continuum coupling. Magnetic field tuning has been proposed to narrow the resonances of a Rydberg series converging to an excited Landau level,⁵ above the ionization limit. Most recently, the Stark-field-ionization resonances in sodium have been shown to narrow near a crossing and have been described quantitatively by WKB quantum defect theory.⁶ (In these last two cases, the autoionization involves only one electron which uses two degrees of freedom to store the energy.) Stark fields have also been used to produce interference narrowing between barium autoionizing states.⁷

In this Letter we report measurements of the $[5d_{3/2}n_2d_{3/2}]_{J=0}$ autoionizing states of barium which show that the interelectron Coulomb interaction can

be adjusted to inhibit autoionization, by variation of the principal quantum number n in a doubly excited Rydberg series. In this fashion, we have observed 3 orders of magnitude change in the autoionization rate for states whose principal quantum numbers differ only by 10%. Because of the strong spin-orbit interaction of the $5d$ electron, the $[5d_{3/2}n_2d_{3/2}]_{J=0}$ Rydberg states are generally jj coupled and hence are mixtures of $5dn_2d^1S_0$ and $5dn_2d^3P_0$ LS -coupled components. However, because of configuration interaction with the $[5d_{5/2}n_3d_{5/2}]_{J=0}$ series, the 1S_0 and 3P_0 amplitudes of the $[5d_{3/2}n_2d_{3/2}]_{J=0}$ Rydberg states change dramatically with increasing principal quantum number, which leads to a pure 3P_0 state at $n_2=60$. Below the $5d^2D_{3/2}$ ionization limit, only $Ba^+(6s)$ ions are possible, and so the continua are of $6s\epsilon l$ type and no open 3P_0 channel of even parity exists. Because of parity and L conservation, the autoionization of the $[5d_{3/2}60d_{3/2}]_{J=0}$ state is inhibited, which results in a linewidth of less than 6 MHz corresponding to a lifetime $\tau \geq 25$ nsec. To observe these metastable autoionizing states, we have used a stepwise multiphoton excitation technique employing two narrow-band cw dye lasers.

As shown in Fig. 1, we used two cw dye lasers to populate $[5d_{3/2}n_2s]_{J=1,2}$ and $[5d_{3/2}n_2d]_{J=0,1,2}$ autoionizing Rydberg states, using two different excitation schemes. In the first scheme, one ring dye laser excited atoms from the ground state to the intermediate $5d6p^3D_1$ state and a second ring dye laser then excited the $[5d_{3/2}n_2d_{3/2}]_J$ autoionizing states as shown in Fig. 2(a). In the second scheme, a dc discharge populated the metastable $5d6s^1D_2$ state and ring dye lasers were used to populate first the $5d6p^1P_1$ intermediate state and then the final $[5d_{3/2}n_2d_{3/2}]_J$ autoionizing states, as shown in Fig. 2(b). In both schemes the

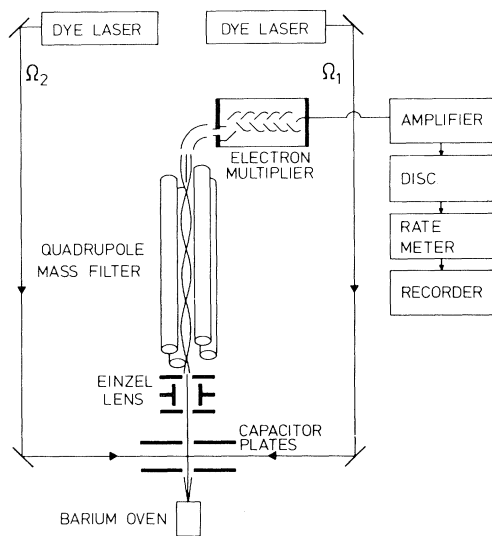


FIG. 1. Experimental setup.

lasers were stabilized to a bandwidth of about 1 MHz with typical power levels of 5 and 100 mW for the two lasers, respectively. The linearly polarized laser beams crossed an atomic beam at 90° and the ions produced by autoionization were focused into the entrance aperture of a quadrupole mass spectrometer by means of an electrostatic lens. The ions were detected with a secondary-electron multiplier. The quadrupole mass filter was especially important to suppress the background ions which were present when the dc discharge was used. A parallel-plate capacitor served to compensate the electric field in the excitation region caused by the electrostatic lens.

When we employed the excitation scheme shown in Fig. 2(a), the dye lasers were operated with stilbene 1 and stilbene 3, respectively. Spectra were obtained by our tuning the frequency of the second dye laser (Coherent model CR 699-29 Autoscan) while the wavelength of the first laser was kept fixed at $\lambda_1 = 413.359$ nm. The broad tuning range (5000 GHz) and narrow bandwidth of the Autoscan made it possible to record narrow as well as broad autoionizing resonances. The total angular momenta J of the Rydberg states were determined by our taking spectra with parallel as well as perpendicular polarizations of the laser beams. To measure term values, a ^{130}Te -vapor cell was used to determine the wavelength of the second laser. For measurements of linewidths the internal frequency scale provided by the wavemeter of the Autoscan was used. The second excitation scheme [(Fig. 2(b)] via the $5d6p\ ^1P_1$ intermediate level was found to be particularly suited to reach $[5d_{3/2}n_2d_{3/2}]_{J=0}$ Rydberg states ($n = 71-84$) of predominantly singlet character. This excitation scheme

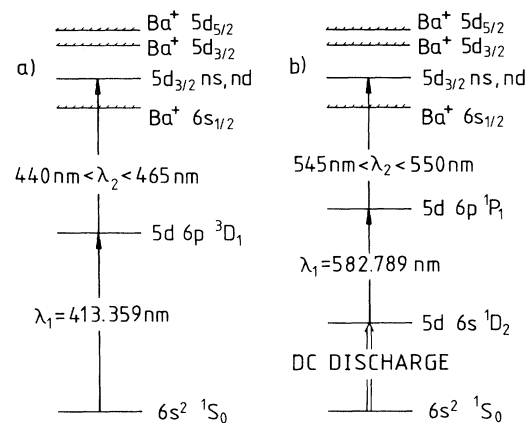
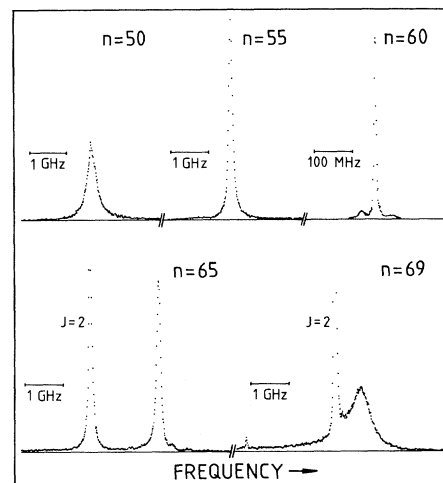


FIG. 2. Excitation schemes.

requires dye lasers operating in the rhodamine 6G and rhodamine 110 spectral range. The first laser was stabilized to the $5d6s\ ^1D_2 \rightarrow 5d6p\ ^1P_1$ transition ($\lambda_1 = 582.789$ nm). The tuning range of the second dye laser (Spectra Physics model 380D) was only 30 GHz. Two marker cavities with free spectral ranges of 150 MHz and 5 GHz provided the frequency calibration.

In Fig. 3 we show the narrowing and broadening of $[5d_{3/2}n_2d_{3/2}]_{J=0}$ autoionizing resonances for principal quantum numbers between $n_2 = 50$ and 69. At higher energies resonances falling within the broad contour of the $[5d_{5/2}14d_{5/2}]_{J=0}$ state are asymmetric and exhibit pronounced Fano-type line shapes (cf. Fig. 4). We summarize our data in Fig. 5 by plotting the normalized linewidths $\Gamma(n_2^*)^3$ versus the effective principal

FIG. 3. Typical line shapes of some $[5d_{3/2}n_2d_{3/2}]_{J=0}$ states. Note the expanded scale at $n_2 = 60$.

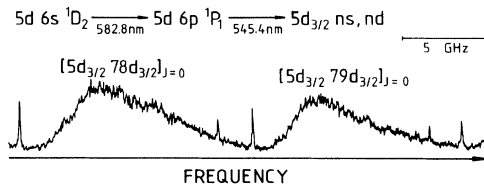


FIG. 4. Broad $[5d_{3/2}n_2d_{3/2}]_{J=0}$ states for $n_2 = 78$ and 79 .

quantum number $n_3^* \pmod{1}$. As usual, n_i^* ($i = 2, 3$) is determined from the binding energy relative to the corresponding ionization limit, so, for example, $E = I_2 - (0.5 \text{ a.u.})/n_2^{*2}$. The three ionization limits used here are the Ba^+ $6s$, $5d_{3/2}$, and $5d_{5/2}$ states, respectively. In Fig. 5, the influence of four perturbing states $[5d_{5/2}n_3d_{5/2}]_{J=0}$, $n_3 = 11$ through 14 , on the $[5d_{3/2}n_2d_{3/2}]_{J=0}$ Rydberg series are compared. To a good approximation all data can be described by one resonance. A decrease in the linewidths from the normal behavior by 3 orders of magnitude is observed. The lowest experimental point corresponds to the $n_2 = 26$ state with a linewidth of 8.5 MHz , slightly larger than that of the $n_2 = 60$ state. Besides $[5d_{3/2}n_2d_{3/2}]_{J=0}$ Rydberg states, $J = 1, 2$ fine-structure components too are narrowed by interference effects. In the remaining part of this Letter, however, we focus our attention on $J = 0$ Rydberg states, because the $J = 0$ channels $[5d_{3/2}n_2d_{3/2}]_{J=0}$, $[5d_{5/2}n_3d_{5/2}]_{J=0}$, and $6s\epsilon s^1S_0$ represent a particularly transparent example of interference narrowing of autoionizing states.

Multichannel quantum-defect theory (MQDT)^{8,9} describes the narrowing of autoionizing states by interference effects implicitly. Recently, configuration

interaction of an autoionizing Rydberg series with a broad interloping resonance has been discussed within the framework of the k -matrix formalism.¹⁰ The same physical situation has been analyzed theoretically,¹¹ by employment of the three-channel quantum-defect theory. Our discussion closely parallels that of Ref. 11.

The bound character of the autoionizing states can be written as a sum of the two jj -coupled $5dnd$ components:

$$|\Psi_b\rangle = \cos\theta |5d_{3/2}n_2d_{3/2}\rangle + \sin\theta |5d_{5/2}n_3d_{5/2}\rangle, \quad (1)$$

where the jj -coupled states are related to LS -coupled states as follows:

$$\begin{aligned} |5d_{3/2}n_2d_{3/2}\rangle &= \left(\frac{2}{5}\right)^{1/2} |^1S_0\rangle + \left(\frac{3}{5}\right)^{1/2} |^3P_0\rangle, \\ |5d_{5/2}n_3d_{5/2}\rangle &= \left(\frac{3}{5}\right)^{1/2} |^1S_0\rangle - \left(\frac{2}{5}\right)^{1/2} |^3P_0\rangle, \end{aligned} \quad (2)$$

and all wave functions are normalized per unit energy, i.e., $\langle 5d_i d | 5d n_i d \rangle = (n_i^*)^3$, so that the wave functions at small r do not depend on n_i at all. This peculiar normalization has a simple classical analog: $\tan\theta$ represents the ratio of the number of revolutions performed in each orbit. Note that this is not the same as the ratio of time spent in each configuration (the usual normalization), since the configurations have different orbital periods. The angle θ defined in Eq. (1) is determined by the coupling of the jj components to each other as well as to the continuum channel. Simultaneously, boundary conditions specific to Coulomb wave functions must be imposed, which requires that θ also depends on the effective quantum numbers of the two jj channels. MQDT^{8,11} summa-

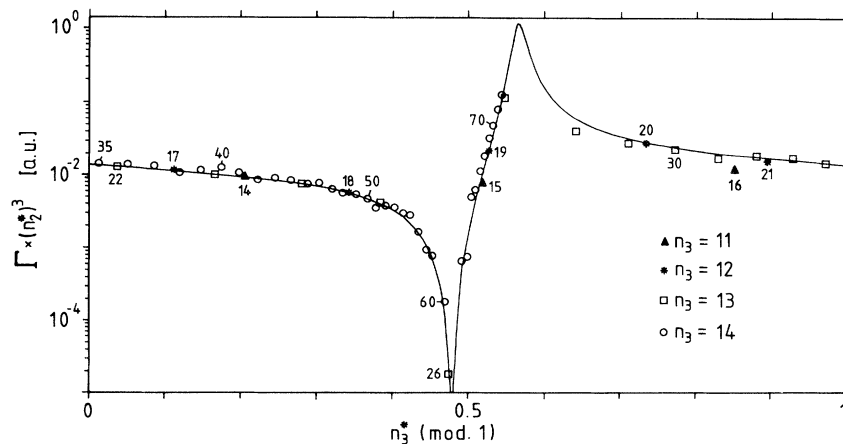


FIG. 5. Measured linewidths of the $[5d_{3/2}n_2d_{3/2}]_{J=0}$ states, with the solid line showing a MQDT fit. The triangles correspond to Rydberg states with $n_2 = 14$ – 16 , perturbed by the $[5d_{5/2}11d_{5/2}]_{J=0}$ resonance. The states with $n_2 = 17$ – 21 , represented by asterisks, are perturbed by the $[5d_{5/2}12d_{5/2}]_{J=0}$ state. The perturber with $n_3 = 13$ affects the Rydberg states with $n_2 = 22$ – 34 , represented by the squares. The circles indicate the states with $n_2 = 35$ – 72 , perturbed by the $[5d_{5/2}14d_{5/2}]_{J=0}$ resonance. Data points at $n_2 = 59, 61, 62$ are missing.

rizes these conditions as follows:

$$\begin{pmatrix} \tan(\pi\tau) & R_{12} & R_{13} \\ R_{12} & \tan\pi(n_2^* + \delta_2) & R_{23} \\ R_{13} & R_{23} & \tan\pi(n_3^* + \delta_3) \end{pmatrix} \begin{pmatrix} A_1 \cos(\pi\tau) \\ \cos\theta \cos\pi(n_2^* + \delta_2) \\ \sin\theta \cos\pi(n_3^* + \delta_3) \end{pmatrix} = 0, \quad (3)$$

where $-\pi\tau$ is the phase shift of the continuum wave function and is obtained by the solving of Eq. (3). A_1^2 is the amount of continuum character, and the R_{ij} values are those defined by Seaton⁸ or Cooke and Cromer¹¹ when the quantum defects δ_i are chosen so that $R_{ij} = 0$. In this formulation, R_{ij} is $1/\pi$ times the sum of the Slater integrals representing the coupling between channels i and j evaluated with the wave functions normalized per unit energy, so that R_{ij} is dimensionless. Note that this requires that $R_{12} = (\frac{2}{3})^{1/2} R_{13}$. In order for Eq. (3) to have a nontrivial solution the determinant of the corresponding matrix has to vanish. By solving Eq. (3) for the phase shift $-\pi\tau$ and taking its derivative with respect to the energy at resonance we obtain

$$\Gamma(n_2^*)^3 = 2\pi^{-1} R_{13}^2 [(\frac{2}{3})^{1/2} \tan\pi(n_3^* + \delta_3) - R_{23}]^2 / [\tan^2\pi(n_3^* + \delta_3) + R_{13}^4]. \quad (4)$$

The expression for Γ was derived by our neglecting the energy dependence of the slowly varying effective quantum number n_3^* . In addition, the small-angle approximation of the tangent was used. In Fig. 5, the solid line represents Eq. (4) with the following parameters: $\delta_3 = 2.435$, $R_{23} = -0.23$, and $R_{13} = -0.17$. These values are very close to those obtained by reduction of the four-channel analysis of Aymar, Camus, and Himdy.¹² As can be seen from Fig. 5, the experimental data are well described by Eq. (4).

Theoretically the autoionization rate Γ vanishes when $\tan\pi(n_3^* + \delta_3)$ equals $R_{13}R_{23}/R_{12} = \frac{3}{2}^{1/2} R_{23}$. At that energy [$n_3^* \pmod{1} = 0.485$], the admixture of the continuum wave function A_1^2 is zero, which thus reduces Eq. (3) to a two-channel MQDT expression

$$\tan\pi(n_2^* + \delta_2)\tan\pi(n_3^* + \delta_3) = R_{23}^2.$$

When this condition is not satisfied, the linewidth is proportional to the mismatch from this condition, evaluated at resonance:

$$\Gamma(n_2^*)^3 = 2\pi^{-1} |\tan\pi(n_2^* + \delta_2)\tan\pi(n_3^* + \delta_3) - R_{23}^2| / R_{12}^2. \quad (5)$$

The states $[5d_{3/2}n_2d_{3/2}]_{J=0}$ with $n_2 = 26$ and 60 corresponding to $n_3^* \pmod{1} = 0.475$ and 0.470, respectively, are close to the point of vanishing resonance width.

In conclusion, the $[5d_{3/2}n_2d_{3/2}]_{J=0}$ autoionizing states of barium provide an illustrative example of how internal interactions can be used, even in very heavy atoms, to enforce simple selection rules that can inhibit autoionization. As a result, $n_2 \approx 80$ states only live for a couple of Rydberg-orbit periods, while the $n_2 = 60$ metastable state lives for nearly 10 000 orbits. Only narrow-band cw dye lasers provide the necessary dynamic range to probe such a large variation in lifetimes.

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