Spectral hole burning in the dielectronic recombination from a continuum of finite bandwidth

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The presence of an electric field converts the intermediate autoionizing Rydberg $n\ell$ states, through which dielectronic recombination passes, into nk Stark states, which have autoionization and capture rates in excess of the radiative decay rates and contribute to dielectronic recombination. In zero field the high ℓ states do not contribute to dielectronic recombination, but the conversion to Stark states makes it possible and raises the dielectronic recombination rate. However, an electric field can also result in coupling to loss channels which locally reduce the dielectronic recombination rate. We have observed holes in the spectrum of dielectronic recombination from the Ba $6p_{3/2}8g$ continuum of finite bandwidth via the intermediate $6p_{1/2}ng$ states. The holes appear when an electric field is applied, and we attribute them to interaction with rapidly decaying $6p_{3/2}8\ell$ states, which diverts flux from dielectronic recombination.

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I. INTRODUCTION

Dielectronic recombination (DR) of an ion and an electron occurs by the capture of the electron, producing an intermediate autoionizing Rydberg state, followed by radiative decay to a bound state of the next lower ionization stage. For example, DR of a ground state Ba^+ ion and an electron occurs in the following way:

$$Ba^+\ 6s + e^-\epsilon\ell' \rightarrow Ba\ 6p_{1/2}n\ell \rightarrow Ba\ 6s_{1/2}n\ell + h\nu. \quad (1)$$

We follow the usual convention that n, ℓ, and m are the principal, angular momentum, and azimuthal angular momentum quantum numbers of the Rydberg electron. In zero field most of the DR is via Rydberg states of low ℓ, since they have large capture rates. The presence of a small electric field can mix Rydberg states of different ℓ, producing Stark states, which together have the same capture rate as the low ℓ states but are more likely to decay radiatively than autoionize. The result is a higher DR rate, as first suggested by Jacobs *et al.* [1]. For much the same reason that electric fields enhance DR rates they are important in other contexts as well. For example, in zero kinetic energy electron (ZEKE) spectroscopy of atoms and molecules electric fields mix optically accessible low ℓ states, which often have high predissociation or autoionization rates, with high ℓ states which have long enough lifetimes that a Rydberg atom or molecule lives long enough to be detected by field ionization [2,3]. Similarly, recent work on doubly excited He $2\ell n\ell'$ states excited by synchrotron radiation has shown that the yield of the $2\ell n\ell' \rightarrow 1sn\ell'$ fluorescence can be dramatically enhanced by the presence of a small field [4]. Again the enhancement is due to the mixing of high ℓ states, which have high fluorescence yields, with the optically accessible low ℓ states, which are more likely to autoionize. Less well appreciated is the fact that electric fields can in some circumstances suppress the DR rate, ZEKE signals, and the fluorescence yield of He $2\ell n\ell'$ states. The suppression occurs when the electric field leads to mixing with rapidly decaying states. In most of these cases the interaction and mixing is between states converging to the same limit. For example, in the presence of a field all the He $2\ell n\ell'$ states of $(-)$ symmetry converging to the He^+ 2ℓ limits disappear from the $2\ell n\ell' \rightarrow 1sn\ell'$ fluorescence spectrum. The disappearance is due to mixing with the rapidly decaying $(+)$ states converging to the He^+ 2ℓ limits [4]. Similarly, the field suppression of the ZEKE spectrum of Ar states converging to the Ar^+ $^2P_{1/2}$ limit is due to mixing of the optically accessible ns' states with the rapidly autoionizing nd' states converging to the same limit [5]. Electric fields suppress the ZEKE spectrum of high ($n > 70$) Rydberg states of NO converging to the lowest rovibrational state of the NO^+ ion [6]. However, in this case a likely cause of the suppression is the field induced coupling between the optically accessible states and rapidly predissociating states converging to higher rotational states of the NO^+ ion, but no sharp spectral features associated with specific predissociating states were observed.

Here we report the observation of spectrally sharp holes in DR from a continuum of finite bandwidth. They are due to the configuration interaction with rapidly autoionizing states converging to a higher ionization limit. We only observe the spectral holes in the presence of an electric field; in the absence of a field coupling to these states is precluded by angular momentum conservation. It is often the case that autoionizing states converging to more than one limit play a role in DR, and in some cases there is evidence of interference [7], but in cases observed to date the effect is purely additive, a clear example being the DR of $Ne^{25+}+e^-$ [8]. The mechanism we propose for the origin of the spectral holes is similar to that suggested by Pratt [6].

This paper is organized in the following way. First we outline the essential ideas, then we describe our experimental approach and present our results and interpretation. Finally, we suggest how this phenomenon might appear in other contexts.

II. PHYSICAL PICTURE

In our experiments we do not study true DR, as expressed by Eq. (1), but DR from a continuum of finite bandwidth, in which the broad Ba $6p_{3/2}8g$ state, which straddles the Ba^+ $6p_{1/2}$ limit, serves as the continuum of finite bandwidth [9,10]. The process we examine is

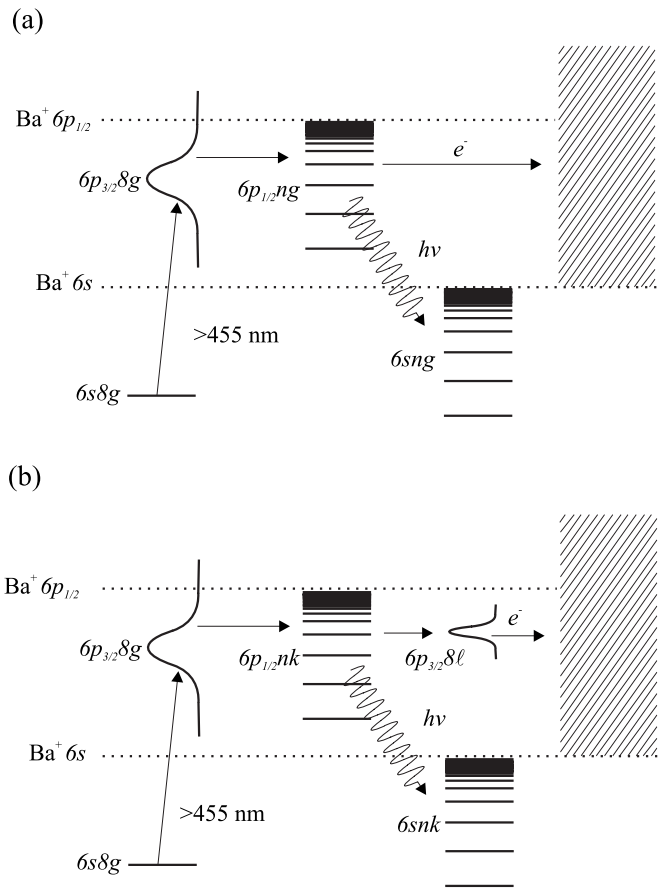

FIG. 1. (a) Schematic diagram of DR from the continuum of finite bandwidth, the Ba $6p_{3/2}8g$ state, in zero field. Atoms are excited to the $6p_{3/2}8g$ by the laser pulse and are captured into the degenerate $6p_{1/2}ng$ state, from which they either autoionize into the continuum or decay radiatively to the bound $6sng$ state, which we detect by field ionization. (b) The same process in the presence of a small electric field. The field converts the $6p_{1/2}n\ell$ states into $6p_{1/2}nk$ Stark states. Capture into them occurs via their $6p_{1/2}ng$ components, and they are coupled to rapidly autoionizing $6p_{3/2}8\ell$, $\ell>4$, states by higher ℓ $6p_{1/2}n\ell$ states. The increased autoionization rate diverts flux from DR.

$$\text{Ba } 6p_{3/2}8g \rightarrow \text{Ba } 6p_{1/2}ng \rightarrow \text{Ba } 6sng + h\nu, \tag{2}$$

which is shown in Fig. 1(a). With a tunable pulsed dye laser we excite atoms to a well defined energy in the $6p_{3/2}8g$ state, the continuum of finite bandwidth, below the Ba^+ $6p_{1/2}$ limit. The atoms have odd parity and well defined angular momentum, $J=3$ or 5 in this work, and both are conserved in zero field. If we think of the $8g$ electron as a classical electron, on each orbit when it comes to its inner turning point it can scatter from the Ba^+ $6p_{3/2}$ core. If the ion is left in its $6s_{1/2}$ state, the electron gains enough energy that autoionization results. However, the scattering can also leave the ion in the $6p_{1/2}$ state, in which case the electron does not gain enough energy to escape but is captured into a $6p_{1/2}ng$ of high n. The $6p_{1/2}ng$ state decays by autoionization or by radiative decay to the bound $6sng$ state. If radiative decay takes place, DR has occurred, and we detect that it has occurred by field ionization of the $6sng$ atom.

The DR process of Fig. 1(a) is altered in several ways by the presence of an electric field of order 10 V/cm, which has no effect on the $6p_{3/2}8g$ state but a substantial effect on the $6p_{1/2}n\ell$ states. The most important effect is that fields in excess of

$$E_g = \frac{2\delta_g}{3n^5} \tag{3}$$

convert the $6p_{1/2}ng$ states into $6p_{1/2}nk$ Stark states, each of which is a superposition of $6p_{1/2}n\ell$ states of all $\ell \geqslant 4$ [11,12]. Here δ_g is the quantum defect of the Ba $6p_{1/2}ng$ states, $\delta_g = 0.02$ [13]. When a Ba atom is excited to the $6p_{3/2}8g$ state in a field it can be captured into all of the $6p_{1/2}nk$ states via their $6p_{1/2}ng$ components, which raises the DR rate by a factor $\sim n$.

Just as the $6p_{1/2}ng$ states are coupled to the $6p_{3/2}8g$ state, $6p_{1/2}n\ell$, $\ell>4$, states can be coupled to autoionizing states converging to a higher limit. In this case the most likely states are 8ℓ, $\ell \geqslant 4$ states, other than the input channels, converging to the Ba^+ $6p_{3/2}$ limit. In zero field the presence of these states is irrelevant due to the conservation of angular momentum, but in the presence of a field they are coupled to the $6p_{1/2}nk$ states via their $\ell>4$ components.

It is straightforward to incorporate the localized loss of DR rate into a calculation of the rate. In zero field the DR rate through a specific $6p_{1/2}n\ell$ state is given by the product of the capture rate and the branching ratio for radiative decay. Explicitly,

$$\Gamma(n\ell) = \frac{\beta R_{n\ell-8g} A_R}{A_R + R_{n\ell-8g} + A_{I_{n\ell}}}. \tag{4}$$

Here $R_{n\ell-8g}$ is the transfer rate from the $6p_{1/2}n\ell$ state to the $6p_{3/2}8g$ state and can be written as $R_{n\ell-8g}=r_{8g}n^{-3}$. By detailed balance $\beta R_{n\ell-8g}$ is the capture rate from the continuum of finite bandwidth. A_R is the radiative decay rate which does not depend on n or ℓ. The denominator of Eq. (4) is the total decay rate of the $6p_{1/2}n\ell$ state. $A_{I_{n\ell}}$ is the autoionization rate of the $6p_{1/2}n\ell$ state into other continua and can be written as $A_{I_{n\ell}}=\gamma n^{-3}$. In an external electric field, Eq. (4) becomes

$$\Gamma(nk) = \frac{\beta R_{nk-8g} A_R}{A_R + R_{nk-8g} + A_{I_{nk}} + R_{nk-8\ell}}. \tag{5}$$

Here $R_{nk-8\ell}$ is the transfer rate from the $6p_{1/2}nk$ state to an $6p_{3/2}8\ell$ state of $\ell>4$. To a first approximation an $n\ell$ state with quantum defect δ_ℓ is uniformly spread over the Stark manifold for electric fields in excess of $E_\ell \geqslant \frac{2\delta_\ell}{3n^5}$. Therefore $A_{I_{nk}}=\gamma_E n^{-4}$, where γ_E is the sum of all the autoionization rates which are mixed into the Stark manifold. Since the field does not affect the $6p_{3/2}8g$ state, the CFB is still only coupled to the $6p_{1/2}ng$ component of the Stark states, and therefore $R_{nk-8g}=r_{8g}n^{-4}$. For the same reason $R_{nk-8\ell}$ exhibits a $1/n^4$ dependence, however because the $6p_{3/2}8\ell$ states are narrow, it also has an approximately Lorentzian dependence. We can then write $R_{nk-8\ell}=r_{8\ell}n^{-4}L(8\ell)$, where $L(8\ell)$ is a normalized Lorentzian with position and width of the $6p_{3/2}8\ell$ state. Equation (5) can now be written as

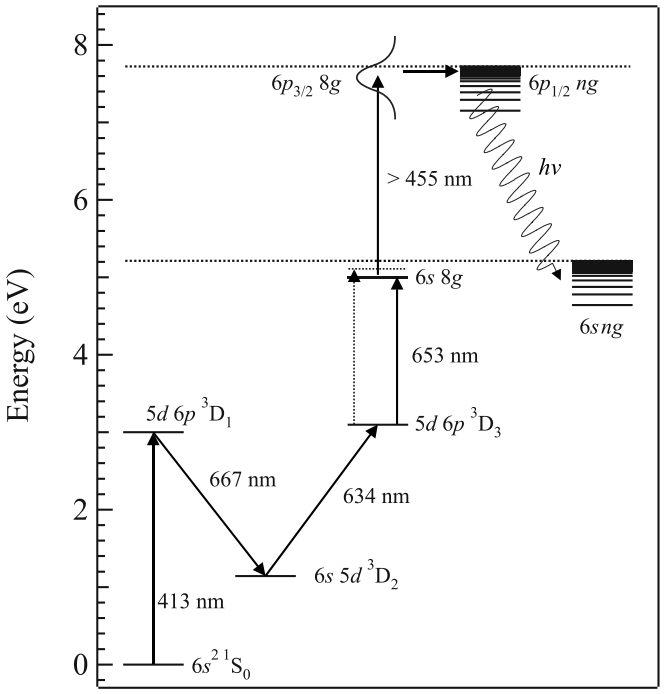

FIG. 2. Schematic diagram of the excitation scheme showing the five transitions driven by the five lasers, the capture by a $6p_{1/2}ng$ state, and the radiative decay to the $6sng$ state.

$$\Gamma(nk) = \frac{\beta r_{8g} A_R}{n^4 A_R + r_{8g} + \gamma_E + r_{8\ell} L(8\ell)}. \quad (6)$$

At the peaks of $R_{nk-8\ell}$ the branching ratio for radiative decay, and thus the DR rate, is substantially reduced.

If the perturbers have a breadth that encompasses a few $6p_{1/2}nk$ states, then they will produce spectral holes in the DR channel with the same widths and energies as the $6p_{3/2}8\ell$ states. A more rigorous treatment of this problem would make use of multichannel quantum defect theory (MQDT). A MQDT analysis would yield similar results to our simple model. The center of the hole would still be within the width of the perturbing state but shifted slightly in energy, and the width of the hole would be comparable to the width of the perturber [14]. In Sec. IV we show that our simple model describes the holes we observe fairly well, however, our data suggest that the interactions between these states is more complicated than our simple model suggests.

III. EXPERIMENT

The apparatus is described elsewhere [11,15], so its description here is brief. Ba atoms in a thermal beam pass along the axis of a set of four rods where they are excited using the multistep laser excitation scheme shown in Fig. 2. Four fixed frequency narrow-band pulsed dye lasers are used to excite the atoms from the ground $6s^2\ ^1S_0$ state to either the $6s8g\ ^3G_4^{16}$ or $^1G_4$ state, with term energies 40 299.384 and 40 300.132 cm^{-1}, respectively [16]. The singlet and triplet designation of these states is tentative. The fifth laser drives the $Ba^+\ 6s \rightarrow 6p_{3/2}$ transition, exciting the atoms to one of the doubly excited $6p_{3/2}8g$ autoionizing states. After the laser excitation, the autoionizing $6p_{3/2}8g$ atoms either autoionize or undergo the quadrupole transition to the $6p_{1/2}ng$ state. The doubly excited Ba $6p_{1/2}ng$ autoionizing atoms either autoionize or decay radiatively via the dipole transition Ba $6p_{1/2}ng \rightarrow 6sng$. In the latter case, the DR process is complete, and we detect that it has occurred by using a 1-kV/cm field pulse to ionize the bound $6sng$ atoms. The resulting electron signal is amplified by a set of microchannel plates and then recorded with a gated integrator as the frequency of the fifth laser is scanned over many shots of the laser.

All five lasers used in our experiment have typical temporal widths of 10 ns and bandwidths of 10 GHz. All five lasers are colinearly polarized in the direction in which the electric field is applied. Only $m=0$ states are excited, and we excite $6p_{3/2}8g$ states of $J=3$ and 5, since the $6p_{3/2}8g$ state is not affected by the small fields we use. In general, however, the field destroys J as a quantum number. As suggested by Fig. 2, there are sources of electrons other than the field ionization of the $6sng$ state atoms, resonant two-photon ionization by the first laser, photoionization from the intermediate $5d6p$ states by the fifth laser, and autoionization of the $6p_{3/2}8g$ state. These processes produce free electrons during the laser pulse, and their presence affects the DR signal. We minimize the effect of two-photon ionization by keeping the power of the first laser as low as possible and its linewidth as narrow as possible. Ionization of atoms in the short lived $5d6p$ states by the fifth laser is minimized by inserting a 20-ns delay before the fifth laser. The number of autoionization electrons from the $6p_{3/2}8g$ state is proportional to the DR signal, so simply reducing it is not useful. To minimize the effect of these free electrons we delay the field ionization pulse until 2 μs after the laser pulse. In addition, we apply the static electric field so as to direct any free electrons away from the detector.

IV. OBSERVATIONS

To show the extent of the continuum of finite bandwidth we show the excitation spectra of the $6p_{3/2}8g$ states starting from the $6s8g\ ^3G_4$ and $^1G_4$ states. These spectra were obtained by collecting the electrons from autoionization as the frequency of the fifth laser is scanned. In Fig. 3(a) we show the spectrum obtained when starting from the $^3G_4$ state, and in Fig. 3(b) we show the spectrum obtained when starting from the $^1G_4$ state. Both spectra exhibit depletion broadening, so the breadth of the states in these spectra exceeds their actual breadth [17]. The $6p_{3/2}8g$ states are best described in jK coupling, in which j, the total angular momentum of the Ba^+ core, is added to the ℓ of the outer electron to produce the angular momentum K. The spin of the $8g$ electron is added to K, yielding the total angular momentum J. The dominant splitting of the $6p_{3/2}8g$ state is due to the quadrupole interaction, which leads to splittings of order 10 cm^{-1} between states of different K [13]. For each value of K there are two possible values of J, but we only excite $J=3$ and 5 states. Presumably each even J state is not far removed in

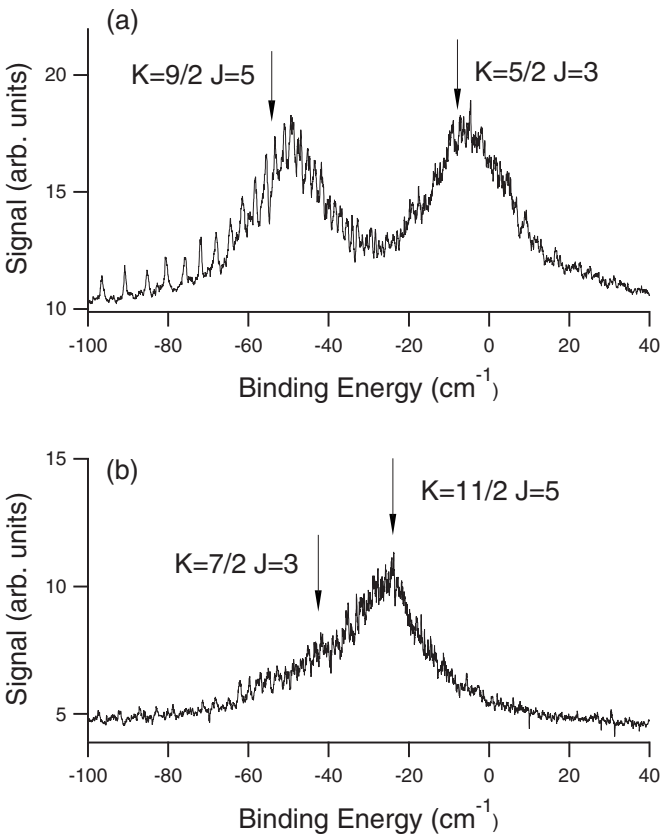

FIG. 3. (a) Excitation spectrum from the $6s8g\ ^3G_4$ state to $6p_{3/2}8g$ $J=3$ and $J=5$ state of $K=7/2$ and $11/2$, respectively. (b) Excitation spectrum from the $^1G_4$ state to $6p_{3/2}8g$ $J=3$ and $J=5$ state of $K=5/2$ and $9/2$, respectively. Both spectra are depletion broadened, so the states appear twice as wide as they actually are.

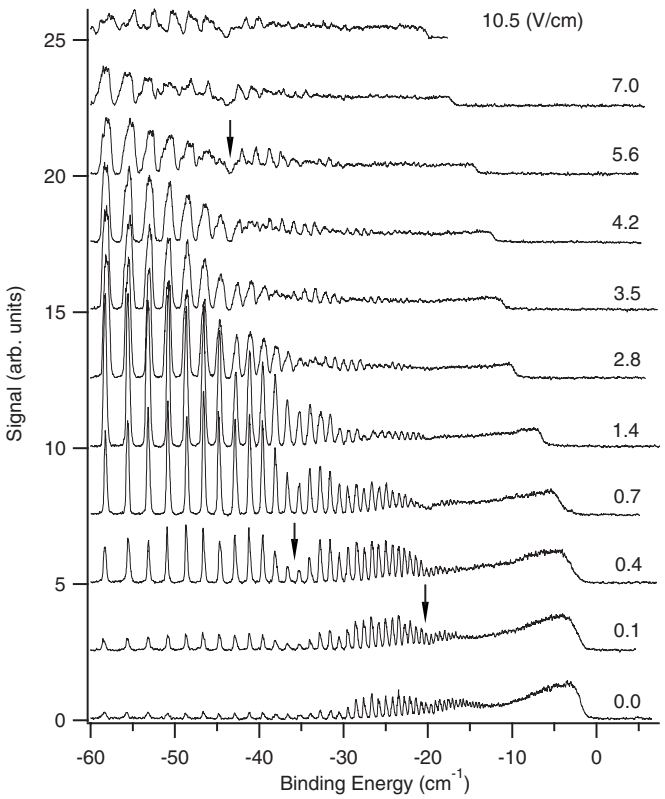

FIG. 4. The DR spectra recorded using excitation from the $6s8g$ $^3G_4$ state in applied fields from 0 to 14 V/cm. We estimate that in zero applied field there is a stray electric field of approximately 70 mV/cm. The depression of the limit by the field is apparent. For example, at 2.8 V/cm it is depressed by 10 cm^{-1}. The spectral holes are indicated by the arrows.

energy from the odd J state of the same K. In Fig. 4 we show DR spectra obtained when starting from the $6s8g\ ^3G_4$ state. The energy scale at the bottom of the figure is relative to the Ba^+ $6p_{1/2}$ limit at 62 296.42 cm^{-1}. Several features are apparent. First, the depression of the limit by

$$W = 2\sqrt{E} \tag{7}$$

due to field ionization is quite apparent. Second, the enhancement of the DR signal due to Stark mixing of the $6p_{1/2}ng$ states for fields in excess of the field of Eq. (3) is clear, as is the decrease when the field exceeds the Inglis-Teller field of $E=1/3n^5$. Present as well, but less apparent, are the spectral holes in the signal which occur at binding energies of 20, 27, 36, and 43 cm^{-1}. They only appear in the presence of the field, and are never very striking, which is why we did not notice them in previous work. In contrast, when we record the DR spectra starting from the $6s8g\ ^1G_4$ state we observe the spectra displayed in Fig. 5, in which the spectral holes are more obvious. The spectral holes occur at the same energies as in Fig. 4, but it is now a straightforward matter to extract their energies and widths, which are tabulated in Table I.

V. INTERPRETATION

As proposed in Sec. II, we attribute the spectral holes to the presence of autoionizing states which converge to Ba^+ limits above the $6p_{1/2}$ limit and are not coupled to the zero field $6p_{1/2}ng$ states due to conservation of angular momentum. The most likely candidates are the Ba $6p_{3/2}8\ell$ states of $\ell \geqslant 4$. In Fig. 6 we present the known and extrapolated energies of the Ba $6p_{3/2}8\ell$ states of $\ell=4-6$. The Ba $6p_{3/2}8g$ energies shown in Fig. 6 are those of the four $J=3$ and 5 states which have been observed [13]. All four values of K are represented, and for each of the observed K states, there is another nearby state of even J. The Ba $6p_{3/2}8h$ energies are taken from the work of Bente and Hogervorst [18], and the $6p_{3/2}8i$ energies are extrapolated from the work of Pruvost *et al.* [19], who measured the energies of the Ba $6p_{3/2}n\ell$ states of $n=11$ and 14 and $\ell \geqslant 4$. Finally, we show the energy of a hypothetical Ba $6p_{3/2}8\ell$ state with a quantum defect of zero. The typical width of the observed $J=3$ and 5 $6p_{3/2}8g$ states is 10 cm^{-1}, the typical width of the $6p_{3/2}8h$ states is 3 cm^{-1}, and the typical width of the $6p_{3/2}8i$ states is 0.5 cm^{-1}. Pruvost *et al.* did not explicitly measure the widths of the $6p_{3/2}n\ell$ states, but the widths of the $6p_{3/2}ni$ states in their spectra are broader than their experimental resolution and are identical to the widths of the $6p_{1/2}ni$ states. The width of the latter was explicitly measured by Jones and

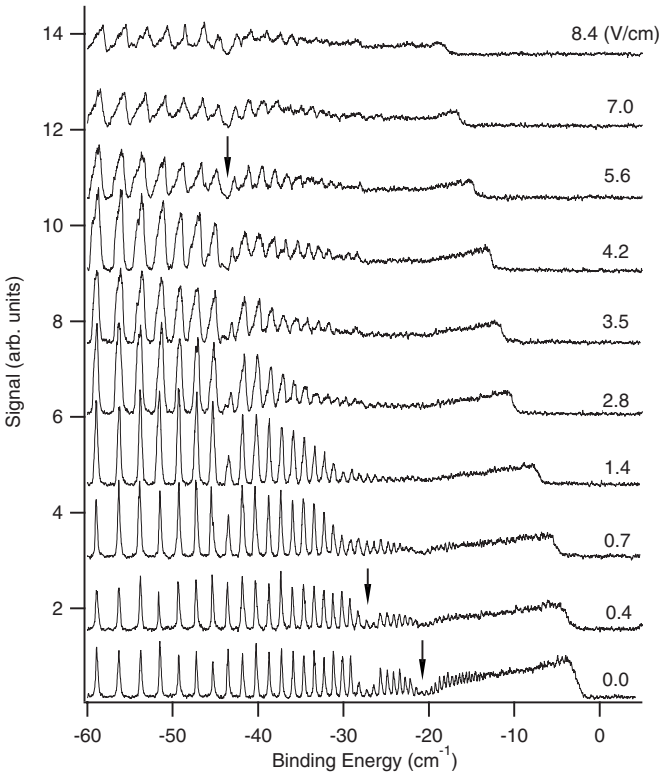

FIG. 5. The DR spectra recorded using excitation from the $6s8g\ ^1G_4$ state in fields from 0 to 8.4 V/cm. We estimate that in zero applied field there is a stray electric field of approximately 70 mV/cm. The depression of the limit by the field is again apparent, and the spectral holes, indicated by the arrows, are more evident than in Fig. 4.

Gallagher for $n=11-13$ [20]. Extrapolating their results yields a width of 0.5 cm^{-1} for the $6p_{3/2}8i$ states. The widths of the higher ℓ states are, of course, narrower [20,21].

Also shown in Fig. 6 are the positions and widths of the spectral holes we have observed. As shown, the widths and spacings of the observed holes match those of the $6p_{3/2}nh$ states, however, each hole is 4–5 cm^{-1} below the energy of the nearby $6p_{3/2}nh$ state. Because the spacings and widths of the holes match those of the $6p_{3/2}nh$ states we believe that interactions with these states are responsible for the holes. If rapid decay simply occurred via the coupling of the $6p_{1/2}nk$ states to the $6p_{3/2}nh$ states as assumed by the model proposed in Sec. II, we would expect the holes to occur at the same energies. Such is evidently not the case, and the interaction of the $6p_{3/2}8h$, $6p_{3/2}8g$, and $6p_{1/2}nk$ states is clearly more complicated than our simple model predicts. A full

TABLE I. Energies and widths (full width at half maximum) of spectral holes.

Term energy (cm^{-1})	Binding energy (cm^{-1})	Width (cm^{-1})
62252.9(6)	43.5(6)	2.8(5)
62259.8(8)	36.6(8)	3.9(8)
62269.2(9)	27.2(9)	3.3(6)
62275.8(9)	20.6(9)	2.7(7)

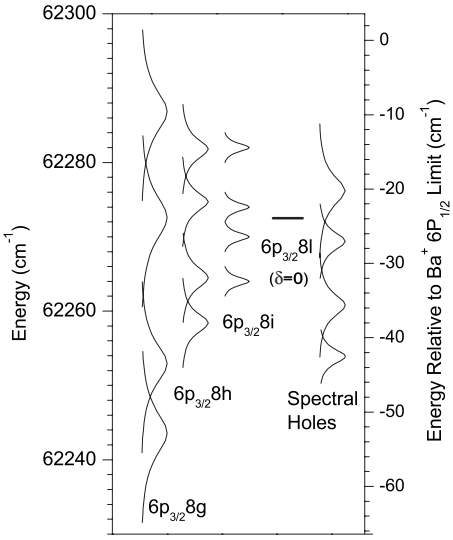

FIG. 6. Energy-level diagram showing the known positions and widths of the Ba $6p_{3/2}\ell$ states of $4\leqslant\ell\leqslant6$, the position of a $6p_{3/2}8\ell$ state of quantum defect 0, and the positions and widths of the observed spectral holes.

MQDT treatment would be in better agreement with our observations because the expected position of the holes would shift to slightly lower energy (~1 cm^{-1}) [14]. However, this small shift is not large enough to improve agreement substantially.

VI. CONCLUSION

Here we report spectral hole burning in DR from a continuum of finite bandwidth. The origin of the holes is the configuration interaction with autoionizing states converging to a higher ionization limit, the Ba^+ $6p_{3/2}$ limit in this case. This coupling only occurs in the presence of an electric field which destroys angular momentum as a good quantum number. This phenomenon should also be observable in other contexts. In ZEKE spectroscopy there should be spectral holes in the Rydberg series due to the coupling with rapidly predissociating states converging to higher rovibrational limits. It should appear in the fluorescence yield spectra of the doubly excited states of He converging to limits above the He^+ $n=2$ limit, where the limits are closer together. For example, low-lying $4\ell n\ell'$ states, which decay rapidly by autoionization, should appear as holes in the $3\ell n\ell'$ fluorescence yield spectrum. Finally, in storage ring experiments it seems quite likely that this phenomenon should be observable in DR of more complex ions that the Li like ions often studied. Promising cases are ions in which there are low-lying excited states which are not easily excited by electron impact of the ground state.

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