Dielectric recombination (DR), the process by which an ion and an electron reconnect via a doubly excited autoionizing Rydberg $nl$ state using electric fields. This notion is based on two essential ideas. First, electric fields enhance the DR rate by Stark-mixing low-$l$ states with high autoionization rates with high-$l$ states with low autoionization rates. Second, the field at which an $l$ state becomes Stark mixed is determined by its quantum defect, a known function of $l$. Consequently, the electric-field dependence of the DR rate should reflect the $l$ dependence of the autoionization rates and thus the contributions of the zero-field $nl$ states to the DR rate. This notion cannot be tested experimentally by examining true DR. However, it can be tested by studying DR from a continuum of finite bandwidth (CFB), for in this case the intermediate Rydberg $nl$ states are restricted to a single value of $l$. Specifically, we have examined the electric-field dependence of DR from two CFB’s, the Ba $6p_{3/2}11d$ and $6p_{3/2}8g$ states. In these two cases the intermediate autoionizing Rydberg states are restricted to the Ba $6p_{1/2}nd$ and $6p_{1/2}ng$ states ($l=2$ and $4$), which have quantum defects of 0.25 and 0.02, respectively. For the same $n$ they are Stark mixed at fields differing by an order of magnitude. We show experimentally that enhancement of the DR rate occurs at fields differing by a factor of 10 for $nd$ and $ng$ states of the same $n$, as expected, confirming that the field dependence of DR can be used to extract information about the contributions of energetically unresolved $l$ states to the zero-field DR rate.

I. INTRODUCTION

Dielectric recombination (DR), the process by which an ion and an electron reconnect via a doubly excited autoionizing Rydberg $nl$ state in a static electric field. It should be possible to separate experimentally the contributions to dielectric recombination (DR) of energetically unresolved intermediate autoionizing Rydberg $nl$ states using electric fields. This notion is based on two essential ideas. First, electric fields enhance the DR rate by Stark-mixing low-$l$ states with high autoionization rates with high-$l$ states with low autoionization rates. Second, the field at which an $l$ state becomes Stark mixed is determined by its quantum defect, a known function of $l$. Consequently, the electric-field dependence of the DR rate should reflect the $l$ dependence of the autoionization rates and thus the contributions of the zero-field $nl$ states to the DR rate. This notion cannot be tested experimentally by examining true DR. However, it can be tested by studying DR from a continuum of finite bandwidth (CFB), for in this case the intermediate Rydberg $nl$ states are restricted to a single value of $l$. Specifically, we have examined the electric-field dependence of DR from two CFB’s, the Ba $6p_{3/2}11d$ and $6p_{3/2}8g$ states. In these two cases the intermediate autoionizing Rydberg states are restricted to the Ba $6p_{1/2}nd$ and $6p_{1/2}ng$ states ($l=2$ and $4$), which have quantum defects of 0.25 and 0.02, respectively. For the same $n$ they are Stark mixed at fields differing by an order of magnitude. We show experimentally that enhancement of the DR rate occurs at fields differing by a factor of 10 for $nd$ and $ng$ states of the same $n$, as expected, confirming that the field dependence of DR can be used to extract information about the contributions of energetically unresolved $l$ states to the zero-field DR rate.

II. THE EFFECT OF AN ELECTRIC FIELD ON DIELECTRONIC RECOMBINATION

In storage ring DR experiments the $n$ but not the $l$ of the intermediate Rydberg states can often be resolved. Here we describe how the field effect provides information about the contributions of energetically unresolved $l$ states using Ba as an example. DR of Ba$^+$ with a free electron via the $6p_{1/2}nl$ states can be thought of as a two-step process, capture, followed by radiative stabilization [12].

\[ \text{Ba } 6s_{1/2} + e^- \rightarrow \text{Ba } 6p_{1/2}nl \rightarrow \text{Ba } 6s_{1/2}nl + h\nu. \]  

(1)

We shall for the moment assume that $m=0$, which implies that the direction of the electron’s motion is the quantization axis. Since capture is the inverse of autoionization, the capture rate can be represented by a constant times the autoionization rate by the principle of detailed balance. After capture, the electron in the Ba $6p_{1/2}nl$ state can either radiatively decay to the bound Ba $6s_{1/2}nl$ state or autoionize into the continuum. Thus, \( \Gamma(nl) \), the contribution to the DR rate of the $6p_{1/2}nl$ state, can be represented by [12].
\[ \Gamma(nl) = \beta A_{nl}(nl) \frac{A_R}{A_{nl}(nl) + A_R}. \]  

Here \( \beta \) is a constant, \( A_{nl}(nl) \) is the autoionization rate of the \( nl \) state, and \( A_R \) is the radiative decay rate from \( 6pnl \) to \( 6snl \). The rate of the radiative decay process of Eq. (1) is independent of \( n \) and \( l \) since it is the \( Ba^+ 6p_{1/2} \rightarrow 6s_{1/2} \) transition with the outer electron remaining a spectator during the process. Inspection of Eq. (2) reveals that, except in cases where \( A_{nl}(nl) = A_R \),

\[ \Gamma(nl) = \beta A_-, \]

where \( A_- \) is the lesser of \( A_{nl}(nl) \) and \( A_R \). To an excellent approximation, \( A_{nl}(nl) = a(l)n^{-3} \), where \( a(l) \) is a rapidly decreasing function of \( l \). Typically, the major contribution to the DR rate comes from low-\( l \) states for which \( A_{nl}(nl) > A_R \), and the total contributions of the \( l \) states of a given \( n \) to the DR rate, \( \Gamma(n) \), can be easily determined by simply counting the states for which \( A_{nl}(nl) > A_R \).

For hydrogenic states any electric field converts the \( nl \) states to \( nk \) Stark states which are linear superpositions of \( nl \) states and, to a first approximation, have autoionization rates equal to the average autoionization rate of all the \( l \) states of the same \( n \) and \( m \), where the quantization axis is the field direction [13–15]. If the average autoionization rate exceeds the radiative rate, then all Stark states contribute to DR, which raises the rate [13,14]. In essence, the field transfers the excess autoionization rates of the low-\( l \) states to the high-\( l \) states. If the radiative decay rate is much higher or lower than the autoionization rates of all the \( nl \) states, then the field has no effect on DR.

Of course, DR occurs only in nonhydrogenic atoms, which have finite-sized cores and \( l \)-dependent quantum defects, resulting in a distinctly nonhydrogenic Stark effect. To be precise, for any given field only the states of \( l > l_E \) are converted to Stark states while those of \( l < l_E \) remain angular momentum states. In other words, the \( l \) states of the lowest \( l \) state converted to a Stark state. We shall shortly provide a more precise definition of \( l_E \). Increasing the field decreases \( l_E \), the boundary for Stark mixing. The onset of field enhancement of the DR rate occurs when \( l_E \) decreases to the value at which an \( l \) state with an autoionization rate exceeding the radiative rate is Stark mixed with higher-\( l \) states, for which the opposite is true. The \( l \) dependence of the quantum defect can arise from the electric multipole moments of an ion core of total angular momentum 1 or greater [16] or from the polarizability of any ionic core [17]. We here restrict our attention to the case in which the quantum defects are due to core polarization since the \( Ba^+ 6p_{1/2} \) ion falls into this category.

To an excellent approximation the binding energy of a nonpenetrating \( nl \) state is given by the hydrogenic energy with a small correction due to the quantum defect [17],

\[ W_{nl} = \frac{1}{2n^2} - \frac{\delta_l}{n^3}. \]

We use atomic units unless specified otherwise. For high-\( l \) states the quantum defect is due to polarization of the ionic core by the Rydberg atom and is given by [17]

\[ \delta_l = n^3 \alpha_d(r^{-4}). \]

where \( \alpha_d \) is the polarizability of the ionic core and \( \langle r^{-4} \rangle \) is the expectation value of the squared field from the Rydberg electron at the ionic core. To a reasonable approximation [18],

\[ \langle r^{-4} \rangle = \frac{1}{n^3(l + 1/2)^5}, \]

so that

\[ \delta_l = \frac{\alpha_d}{(l + 1/2)^5}. \]

In the presence of an electric field the high-\( l \) states, which have small quantum defects, are converted to Stark states, while the low-\( l \) states, which have large quantum defects, are unaffected [19]. Consequently, the field \( E \) defines \( l_E \), the lowest-\( l \) state converted to a Stark state. States of \( l > l_E \) become what we shall here refer to as Stark states, although they are not the parabolic states. States of \( l < l_E \) remain angular momentum states and are unaffected by the field. This notion is depicted graphically in Fig. 1.

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If we assume that states of \( l > l_E \) have negligible quantum defects, we can relate \( l_E \) to \( E \) by equating the energy shift \( \delta_l/n^3 \) to the Stark shift of the extreme Stark state to define \( l_E \) implicitly,

\[ \frac{\delta_l}{n^3} = \frac{3n(n - 1 - l_E)E}{2}. \]

For \( l_E \ll n \) this reduces to
where \( E_{IT} \) is the Inglis-Teller field \( 1/3n^5 \).

If we know the autoionization rates and quantum defects as a function of \( l \) it becomes straightforward to calculate the DR rate as a function of the field. In the field \( m \) remains a good quantum number, and for small fields \( n \) does as well. We can write the recombination rate in the field \( E \) through all states of a given value of \( n \) and \( m \) as

\[
\Gamma_E(n,m) = \beta \sum_{l=|m|}^{l_e-1} \frac{A_{nl}(nl)}{A_{nl}(nl) + A_R} + \frac{(n-l_E)}{\bar{A}_s(E)A_R} \frac{\bar{A}_s(E)A_R}{\bar{A}_n(E)A_R}.
\]

If all \( m \) states make equal contributions to the DR rate, then summing the contributions for all \( m \) states yields the contribution of a given \( n \) state to the DR rate vs field. Explicitly,

\[
\Gamma_E(n) = \sum_{m=-(n-1)}^{n-1} \Gamma_E(n,m).
\]

To illustrate the field dependence graphically, we show in Fig. 2 the \( \Gamma_E(25,0) \) as a function of field for a model atom. In this figure, the autoionization rates in atomic units for \( l = 0 \rightarrow 4 \) are 0.09\( n^{-3} \), 0.02\( n^{-3} \), 0.05\( n^{-3} \), 0.31\( n^{-3} \), and 0.05\( n^{-3} \), respectively [20]. The autoionization rates for \( l \geq 4 \) are given by \( A_{nl} = 24e^{-1.5l_{III}n} \), which approximates those of the Ba 6p\( ^1l_2nl \) states [21]. We assume that the quantum defects of \( l \geq 4 \) states are determined from Eq. (7) using a core polarizability of 120\( \alpha_0^2 \), and that the quantum defects of the \( \leq 3 \) states are given by 0.5, 0.3, 0.2, and 0.1, respectively. We have used two values for the radiative decay rate, \( A_R = 3.88 \times 10^{-10} \) and \( 3.88 \times 10^{-8} \). The former is 10% of the Ba 6p\( ^1l_26s \) transition rate, while the latter is ten times it. In both cases there is no field dependence of the DR rate at low field, since only the highest-\( l \) states, which have autoionization rates less than the radiative rate, are affected. The DR rate begins to increase at the field at which the \( l \) state with an autoionization rate in excess of the radiative rate joins the Stark manifold. For \( A_R = 3.88 \times 10^{-10} \) the increase in the DR rate begins at \( l=10 \) and \( E=0.4 \) V/cm, and for \( A_R = 3.88 \times 10^{-5} \) it begins at \( l=7 \) and \( E=1.5 \) V/cm. There is no field dependence at fields at or above the Inglis-Teller field since the Stark mixing is complete. In an experiment, the observed DR rate is a sum over all \( m \) states, with a weighting dependent on the experimental geometry. However, even if we assume an equal weighting of \( m \) states, and compute \( \Gamma_E(25) \) as a function of field for \( A_R = 3.88 \times 10^{-10} \) and \( 3.88 \times 10^{-8} \), the field enhancements occur at the same places as in Fig. 2, because \( l_E \) does not depend on \( m \).

Figure 2 demonstrates the essential point, that the \( E \)-field dependence of the DR rate should reflect the \( l \) dependence of the quantum defects and autoionization rates. Consequently, this technique should be useful in analyzing the contributions to DR of energetically unresolved \( l \) states in storage ring experiments.

III. DIELECTRONIC RECOMBINATION FROM A CONTINUUM OF FINITE BANDWIDTH

In true DR there is no control of the impact parameter of the electron-ion collisions leading to DR, and hence no control over the \( l \) of the Rydberg electron in the intermediate autoionizing state. Consequently, it would be impossible to verify experimentally the notions outlined above by studying true DR. Instead, we have replaced the true continuum with a continuum of finite bandwidth (CFB) [11], allowing us to select the \( l \) of the Rydberg electron in the intermediate autoionizing state by choosing different continua of finite bandwidth. In this experiment, either of the two broad autoionizing Ba* 6p\( ^3l_21d \) or Ba* 6p\( ^3l_28g \) states converging to the Ba* 6p\( ^3l_2 \) limit serves as the continuum of finite bandwidth. Specifically, we have studied the two processes

\[
{\text{Ba}6p_{3/2}1d} \rightarrow {\text{Ba}6p_{1/2}nd} \rightarrow {\text{Ba}6snpd + hv}
\]
between the 6s2 ground state to the 6p3/2 11d state, which is the continuum of finite bandwidth. From it, the 11d electron can either autoionize into the true continuum or be captured into the degenerate 6p1/2 nd state (represented by the horizontal arrow). If capture occurs, the 6p1/2 nd state can either autoionize or decay radiatively to a stable 6snd state (represented by the solid arrow). In the latter case, dielectronic recombination has occurred and is detected by field ionization of the 6snd Rydberg state.

The internal transition from the 6p3/2 11d(8g) → 6p1/2 nd(ng) state is analogous to capture in true DR. This transition is a quadrupole transition, whereas in true DR it is a dipole transition, an insignificant difference for electron scattering. The 11d(8g) state is like a storage ring, and each 11d(8g) electron collides with the Ba 6p3/2 core roughly 20 times before autoionization occurs. The most important difference between true DR and DR from a CFB is the one noted above. In true DR the electron-ion collisions have all possible impact parameters, and the autoionizing Rydberg states resulting from capture of the electron can be in all nl states. In contrast, in DR from a CFB there is only one l value for the entrance channel. The fact that there is only one entrance channel is crucial for this experiment, for it allows us to test our earlier claim that l states with different quantum defects join the Stark manifold, and therefore begin to contribute to the enhancement of DR, at different fields. In this particular experiment we are taking advantage of the difference between true DR and DR from a CFB, i.e., the fact that we can control which low-l state is the entrance channel. It is, however, worth bearing in mind the essential similarity of true DR and DR from a CFB for the study of field effects. In true DR the low-l states have the largest capture rates, so, irrespective of which low-l state we choose as the entrance channel using the CFB, the result is qualitatively the same as for true DR.

For our present purposes the most important difference between the 6p1/2 nd and 6p1/2 ng states is the difference in their quantum defects. Specifically, \( \delta_2 = 0.25 \) and \( \delta_1 = 0.02 \), so for a given \( n \) the electric fields at which these states become Stark states should be different by an order of magnitude, as discussed in the previous section. If the notions presented in the previous section are correct, for a given value of \( n \) the onset of field enhancement of DR should occur at fields differing by the same factor.

### IV. EXPERIMENTAL APPROACH

Ba atoms are excited sequentially to the continua of finite bandwidth, either the 6p3/2 11d state or the Ba 6p3/2 8g state, using the isolated core excitation approach. The 6p3/2 11d state is created using three 5 ns Littman dye lasers as shown in Fig. 3(a). The first two lasers are fixed in frequency to drive the transitions

\[
6s^2 1S_0 \rightarrow 6s6p 1P_1 \rightarrow 6s11d 1D_2.
\]

The third laser drives the transition from the 6s11d to the 6p3/2 11d state, and its frequency is scanned near the Ba\(^+\) 6p1/2 limit. All three laser pulses overlap in time and space. In essentially the same manner, the 6p3/2 8g state is created using five 5 ns Littman dye lasers as shown in Fig. 3(b). The first four lasers are fixed in frequency to drive the transitions

\[
6s^2 1S_0 \rightarrow 5d6p 3D_1 \rightarrow 6s5d 3D_2 \rightarrow 5d6p 3D_3 \rightarrow 6s8g 3G_4.
\]
Q-switched Nd:YAG (yttrium aluminum garnet) laser running at a 20 Hz repetition rate.

Aside from the differing excitation schemes, the experiments from the two continua of finite bandwidth are conducted in the same way. As shown in Fig. 4 a thermal beam of barium atoms effuses from a resistively heated oven and passes down the axis of a set of four rods 0.24 cm in diameter and 1.9 cm apart. The lasers counterpropagate to the atomic beam and overlap the beam along the axis of the rods. A fixed voltage can be placed on the rods to create a static field during the excitation. Typically, the polarization of the lasers is parallel to the field so as to excite the field in the continua of finite bandwidths. In Fig. 5 we show DR under the same conditions from the 11d CFB. In Fig. 5(b) we show the DR signals obtained from the 8g CFB with no static field and in the presence of a 1.5 V/cm static field, and in Fig. 5(b) we show DR under the same conditions from the 11d CFB. In Fig. 5 we see that in the 1.5 V/cm traces there is a substantial increase in DR from both CFB’s as compared to the zero-field traces; however, it is obvious that enhancement is strikingly different depending on whether DR occurs from the 8g or 11d CFB, i.e., through l=4 or 2 intermediate $6p_{1/2}nl$ states. In the 1.5 V/cm traces the enhancement of DR extends to a far lower binding energy for the 8g CFB than for the 11d CFB. This observation is what we expected to see. What we did not expect to see is that the enhancement of DR from the 8g CFB seems to disappear at the energy at which the enhancement of DR from the 11d CFB begins to appear.

From Fig. 5 it is apparent that DR via the $6p_{1/2}ng$ states is enhanced at lower field than via the $6p_{1/2}nd$ states of the same n. To make accurate measurements of the fields at which the enhancements occur, we have fixed the frequency of the final laser on the zero-field $6p_{1/2}2nl$ state and scanned the field in which DR occurs. In Fig. 6 we have plotted the fields at which 50% of the enhancement of the DR rate occurs, vs $1/n^5$ for both the nd and ng states. The enhancement fields for the nd and ng states are given by $0.34(1)n^{-5}$ and $0.033(1)n^{-5}$, respectively. In other words, we observe the expected $n^{-5}$ behavior and an order of magnitude difference between the fields for the d and g states, but our observed values do not agree with the values predicted from Eq. (9), which are $0.16n^{-3}$ and $0.013 n^{-5}$. In both cases the enhancement fields are a factor of 2 higher than predicted by Eq. (9), and the discrepancy is due to simplifying assumptions used for beam overlap. In Fig. 4 a thermal beam of barium atoms effuses from a resistively heated oven and passes down the axis of a set of four rods 0.24 cm in diameter and 1.9 cm apart. The lasers counterpropagate to the atomic beam and overlap the beam along the axis of the rods. A fixed voltage can be placed on the rods to create a static field during the excitation. Typically, the polarization of the lasers is parallel to the field so as to excite the continua of finite bandwidths. In Fig. 5 we show typical scans of the final laser wavelength when observing DR with and without a static field present. The traces presented in Fig. 5 have been normalized to the autoionization signals from their respective continua to remove the differences in the DR signals due to the different energy variations in the number of atoms excited to the two continua of finite bandwidths.
in deriving Eq. (9). Let us consider the \( nd \) states first. The enhancement occurs not at \( E=0.16n^{-5} \) but at the Inglis-Teller field. Why this occurs is easily understood by examining the field–energy-level diagram for the Ba \( 6p_{1/2}nl \) states shown in Fig. 7. As shown, the \( nd \) state does not have a field-independent energy until it intersects the Stark manifold but is strongly repelled by the \( (n+1)p \) state and only intersects the Stark manifold at the Inglis-Teller field. The observed enhancement field for the \( ng \) states is also a factor of 2 higher than that predicted by Eq. (9), for a similar reason; it has a nonzero Stark shift for fields less than that given by Eq. (9). Although Fig. 6 does not agree perfectly with our simple model, it dramatically illustrates our main point, that the field dependence of the DR enhancement is different depending on the \( l \) state through which DR occurs.

An important effect noted above is the fact that the enhancement of the DR rate in \( ng \) states decreases dramatically at the field at which the enhancement of the \( nd \) states occurs, i.e., at the Inglis-Teller field. Once the field has increased to the field at which the \( ng \) state is mixed into the manifold, further increases in the field do not increase the capture rate but do increase the loss due to rapid autoionization through the acquired \( l<4 \) character, which lowers the DR rate as the field approaches the Inglis-Teller field. DR from the \( 6p_{1/2}11d \) CFB is somewhat simpler than from the \( 6p_{3/2}8g \) CFB, because the Ba \( 6p_{1/2}nd \) states do not join the Stark manifold until the Inglis-Teller field, as is shown by Fig. 6. Since the \( nd \) state does not mix into the manifold until the Inglis-Teller limit, no other \( l \) states can be added to the manifold at a higher field, and the decline in the DR rate noted in the \( g \) states cannot occur. Stated another way, because the \( nd \) states join the Stark manifold at the Inglis-Teller field, there are only two cases, no Stark mixing and full Stark mixing, as has been shown previously [22,23].

While our focus has been on the electric fields at which the enhancement occurs, quantitative measurements of the enhancements are also useful. By making a series of wavelength scans such as those shown in Fig. 5 we have determined the enhancement factors \( R_{ng}(E) \) for different \( nl \) states as a function of the field. Explicitly, we define the enhancement factor as the ratio of the DR signal in the field to the zero-field signal. In Fig. 8(a) we show the enhancement factors for \( ng \) states of \( 39 \leq n \leq 48 \), and in Fig. 8(b) we show the corresponding enhancement factors for the \( nd \) states.

As shown by Fig. 8 the maximum enhancement factors for the \( ng \) and \( nd \) states are \( R_{ng}(E)=13(3) \) and \( R_{nd}(E)=9(2) \). These values can be compared to estimates for the enhancement factors obtained by following the procedure outlined by Ko et al. [22]. Specifically, we can write the enhancement factor for DR via an \( ng \) state \( R_{ng}(E) \) as the ratio of the DR rate through the \( g \) character of all the \( m=0 \) Stark states (for \( 0.03n^{-5}<E<0.13n^{-5} \)) to the DR rate through the zero-field \( ng \) state. Explicitly,

\[
R_{ng}(E) = \frac{(n-4)\beta R_{ng}(E)A_g[\bar{R}_{ng}(E)+\bar{A}_g(E)+\bar{A}_R]}{\beta R(ng)A_g[R(ng)+\bar{A}_{Ai}(ng)+\bar{A}_R]}. \tag{16}
\]

Here \( \beta R(ng) \) is the capture rate into the \( 6p_{1/2}ng \) state from the \( 6p_{3/2}8g \) state (the constant \( \beta \) was defined previously),
It can be further simplified by using the Stark states, leading to that factor in the numerator. Since Stark state and $A_{ng}$ and $A_{nf}$ continue. We use $(\alpha)_{n}=0.03$, and the value $a_{n}(E)=0.035$, appropriate for $0.013n^{-3}<E<0.13n^{-5}$ [20,24], we find that, for $n=45$, $R_{ng}(E)=31$, which is a factor of $2^{3}$ larger than our measured value. For DR through the $6p_{1/2}n\ell$ states we use the values $r_{\ell}=0.03$, $a_{n}=0.06$, and $a_{n}(E)=0.53$ [20,25], appropriate for fields in excess of the Inglis-Teller field. With these values and $n=45$ we find $R_{nd}(E)=7$, in reasonable agreement with our measured values.

We attribute of the discrepancy between the calculated and measured $ng$ enhancement to partial Stark mixing of the $nf$ states into the Stark manifold. By the time the $ng$ states have joined the Stark manifold, the $nf$ states have partially joined it, sharing their large autoionization rates with the Stark states and suppressing the DR rate. If the field is large enough to completely admix the $nf$ states to the Stark manifold the value of $a_{n}(E)$ is not 0.035, but 0.345, and $R_{ng}(E)=6$. Mixing only 30% of the $nf$ state into the Stark manifold leads to $a_{n}(E)=13$ and $R_{nd}(E)=13$, in agreement with our observations.

VI. CONCLUSION

We have observed a substantial difference in the behavior of DR from a CFB in a static field depending on the $l$ state through which recombination occurs. We have shown that the field at which the DR rate enhancement begins is ten times lower when recombination occurs through the $g$ states than it is when recombination occurs through the $d$ states. We have developed a simple model that agrees reasonably well with the observed results, as well as a more sophisticated calculation which also reproduces our experimental results. These experiments show that by using $E$ fields it should be possible to extract information about the $l$ dependence of DR rates, even though the high-$l$ states are energetically unresolved.

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