Mg $3pnk$ autoionizing states in electric fields

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We have measured the transitions from the bound $3snk$ to the autoionizing $3pnk$ Mg Stark states in static and microwave electric fields. Due to the small separation of the Mg^+ $3p_{1/2}$ and $3p_{3/2}$ states there is substantial configuration interaction between Stark states converging to the two limits, and the n^{-4} scaling of the autoionization widths expected from the simplest one-limit picture is not observed.

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I. INTRODUCTION

As described by Jacobs, Davis, and Kepple [1,2] an electric field has two effects on autoionizing Rydberg states. First, it lifts the near degeneracy of the nl states, converting them to the nk Stark states which have linear Stark shifts. Second, it alters the autoionization rates from their zero field values. Here n, l, k, and m are the principal, orbital angular momentum, Stark, and azimuthal angular momentum quantum numbers. In a field the good quantum numbers are n, k, and m. The linear Stark energy shifts of autoionizing states were first observed by Freeman and Bjorklund [3], and more recently by Armstrong *et al.* [4].

The Stark states have autoionization rates which fall between the high rates of the low-l states and the very small rates of the high-l states. For a given n and m the autoionization rates for all k states are typically of the same order of magnitude and are often given approximately by the average autoionization rates of the l states of the same n and m [2,5]. In contrast, the zero-field nl states have autoionization rates which drop by orders of magnitude as l is increased form zero to $n-1$ [6]. The autoionization rate of a Stark state is proportional to its low-l character, which falls as $1/n$ with increasing n. This dilution of the low-l character with increasing n, combined with the overall $1/n^3$ scaling of the zero-field autoionization rates, suggests a $1/n^4$ scaling for autoionization rates in electric fields. The $1/n^4$ scaling has been observed in Ba $6p_{1/2}nk$ autoionizing Rydberg states by Jaffe *et al.* [7] and Jones and Gallagher [8].

The substantial effect of an electric field on autoionization rates is important for dielectronic recombination, the recombination of an ion and an electron via an autoionizing state [1,2]. Those autoionizing states with autoionization rates exceeding their radiative decay rates contribute equally to the total dielectronic recombination rate. In zero field it is often the case that the low-l states have autoionization rates which are orders of magnitude in excess of the radiative decay rates, while the high-l states have autoionization rates below the radiative decay rates and do not contribute to dielectronic recombination. In a field the l states are mixed and converted to Stark states, all of which have autoionization rates exceeding the radiative decay rate. Thus all the Stark states contribute to the dielectronic recombination rate, and as a result, the recombination rate increases in a field [1,2].

The first experimental suggestion of the effect of electric fields on dielectronic recombination rates was the measurement of Belić *et al.* of the dielectronic recombination of $Mg^+ + e^-$ [9]. They observed rates in a field a factor of five higher than the rates calculated in zero field [10]. Later calculations, including the field effects, removed much of this discrepancy in the total cross section [11–14]. To provide a more complete experimental picture Muller *et al.* measured the final-state distribution of the Mg Rydberg atoms resulting from dielectronic recombination [15,16]. These measurements have shown that, while the later calculations gave reasonable values for the total cross sections, they did not give the experimentally observed final-state distributions in all cases.

Here we report the investigation of the effects of electric fields on the autoionization rates of the Mg $3pnk$ Stark states in static and 13 GHz microwave fields. These measurements were undertaken to complement the dielectronic recombination measurements by providing more detailed information about the field's effect on isolated atomic states. We expected to observe results very similar to those obtained in Ba, and, while there are similarities, there are striking differences as well. In the sections which follow we describe the experimental procedure, present the results, and contrast these observations in Mg with previous observations in Ba.

II. EXPERIMENTAL APPROACH

These experiments are based on multistep laser excitation of Mg atoms in a beam [17,18]. Since the apparatus and approach are almost identical to those used previously, we give only a brief description [18]. We use a resistively heated oven to produce a beam of Mg atoms. Ap-

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proximately 20 cm from the oven the beam passes through the interaction region where there is a static or microwave electric field. In the interaction region the Mg atoms are excited from the ground state to the 3*pnk* autoionizing states by three pulsed dye laser beams as shown in Fig. 1, an energy level diagram for Mg. The wavelengths of the first two lasers are fixed to the Mg $3s^2 \rightarrow 3s3p$ and $3s3p \rightarrow 3snk$ transitions at 285 and ~376 nm (all states are singlet states) to excite the ground-state atoms to the bound 3*snk* Rydberg Stark state. The wavelength of the third laser is scanned over the $3snk-3pnk$ transition while detecting Mg^+ ions from the decay of the 3*pnk* state. Since there is negligible direct photoionization of the 3*snk* state into continua above the Mg^+3s limit, the third laser only excites the inner 3*s* electron to the 3*p* state. The outer *nk* electron is only a spectator. Since the power of the third laser is kept low enough that the observed signal is linear in the laser power, the ion signal is proportional to the photoexcitation cross section of the 3*pnk* state.

For the static-field measurements the interaction region was defined by a pair of aluminum field plates spaced by 0.462 cm. Voltages of up to 3400 V were placed across the plates leading to fields as high as 7360 V/cm. In these experiments the laser beams, the atomic beam, and the electric field direction were mutually orthogonal. The lasers were polarized in the direction of the electric field.

In the microwave field experiments the interaction region was inside a microwave cavity [19]. The cavity was a piece of X band waveguide 20 cm long closed at both ends. The waveguide contained a septum to allow application of a static field in the same direction as the microwave field. The static field enabled us to extract the Mg^+ product of the decay of the 3*pnk* states from inside the cavity, and the presence of the septum had negligible effect on the TE_{10n} cavity modes. The cavity was operated on the TE_{109} mode at 12.85 GHz. The atomic beam passed into and out of the cavity through 1 mm diameter holes in the sidewalls of the cavity, passing 1 mm above the septum inside the cavity. The laser beams were counter propagating to the atomic beam and thus excited a pencil shaped volume of atoms inside the cavity. There is a 1 mm diameter hole in the center of the top of the cavity, and only those ions formed directly below the hole are expelled form the cavity when a voltage pulse is applied to the septum. Consequently, we only detect ions from atoms at the antinode of the microwave field in the center of the cavity.

The three dye lasers are pumped by the second harmonic of a continuum QC661 Q switched neodymium-doped yttrium aluminum garnet (Nd:YAG) laser running at a 20-Hz repetition rate. The first and second dye lasers are Littman-type oscillators followed by single stage amplifiers. These lasers produce 5 ns pulses with 1 mJ energies and 1 cm^{-1} linewidths. These visible pulses are doubled in angle tuned potassium dihydrogen phosphate (KDP) crystals to produce 50–100 μJ pulses at 285 and 376 nm, as shown in Fig. 1. The third dye laser is a Quanta Ray PDL1 dye laser which produced slightly higher pulse energies at 560 nm and was sometimes used with an intracavity étalon to provide a linewidth of 0.1 cm^{-1}. When doubled to 280 nm in a KDP crystal the linewidth was 0.13 cm^{-1}. The wavelength of the third laser was measured by recording transmission fringes through a 0.67 cm^{-1} free spectral range étalon.

III. OBSERVATIONS

Before we could study the 3*pnk* autoionizing Stark states we had to locate the bound 3*snk* Stark states. We did so by scanning the wavelength of the second laser while keeping the third laser tuned near one of the Mg^+ resonance lines. This process was repeated for many values of the static field, the procedure used by Zimmerman *et al.* to map the Stark spectra of the alkalis [20]. We mapped the 3*snk* Stark energy levels from $n = 11$ to 19. The field dependence of the bound Stark manifold states is shown qualitatively by Fig. 2. At static fields where the levels are well resolved there are evenly spaced Stark levels, which we label as Stark *k* states, and an interloping state, which we label as *i* and identify as origi-

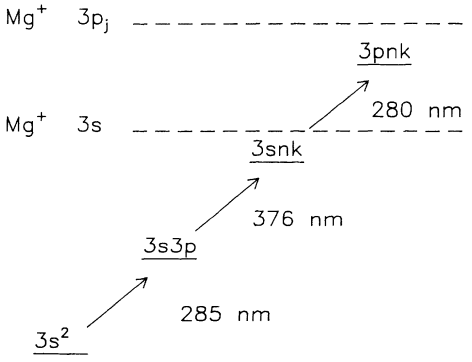

FIG. 1. Energy-level diagram for the excitation of the Mg 3*pnk* Stark states.

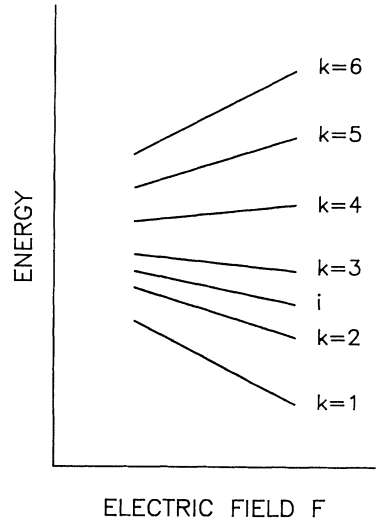

FIG. 2. Qualitative energy level dependence of the bound Mg Stark states showing the regularly spaced Stark states, labeled $k = 1-7$, and the interloper state labeled *i*.

nating from a p state. Not surprisingly, the spectra from these two kinds of states are different.

Although it may seem peculiar to do so, we begin by considering the spectra from the interloper states. As a typical example, in Fig. 3 we show the spectra from the interloper in the $n=14$ Stark manifold as a function of the frequency of the third laser. There are two groups of peaks in the spectrum centered approximately at 35669 and 35761 cm^{-1}, the $Mg^+ 3s_{1/2}-3p_{1/2}$ and $3s_{1/2}-3p_{3/2}$ transition frequencies [21]. Within each group the lines are spaced by approximately 6.5 cm^{-1} the hydrogenic $n=14$ Stark state splitting at a field of 3650 V/cm. Consequently we assign the final states of the two groups of lines of Fig. 3 as the $n=14$ Stark states converging to the $Mg^+ 3p_{1/2}$ and $3p_{3/2}$ limits. This assignment is supported by the observation that the spacing between the lines is approximately linear in the electric field, as expected for Stark states. Why is it possible to observe all the autoionizing $3pnk$ Stark states from the interloper state? Without carrying out detailed calculations it is difficult to be certain, but it is possible to give a likely explanation. The fact that the bound interloper does exhibit a Stark shift suggests that it has acquired Stark character, with more from Stark states lying nearer than from those lying farther away. The admixtures of the bound Stark states in the interloper allow all of the autoionizing $n=14$ Stark states to be excited as shown by Fig. 3. Of course mixing in the final states can also lead to a spectrum as shown in Fig. 3.

If we examine the spectra from the regular bound Stark states we observe somewhat simpler spectra. The results observed for the transitions from the $3s18k$ states to the $3p_{1/2}18k$ states are typical. In Fig. 4 we show the spectra obtained from different $3s18k$ Stark states in a field of 670 V/cm. With the convention of Fig. 2, the lowest energy member of a Stark manifold of $m=0$ has the Stark quantum number $k=1$ and the highest member $k=n$. As shown by Fig. 4, the lower and upper numbers of the Stark manifold lead to single transitions from the $3s18k$ to the $3p_{1/2}18k$ states. However, the inner members of the $n=18$ Stark manifold lead to multiple peaks. Why the difference exists can be appreciated by examining Fig. 5, an energy-level diagram of the $3p_{1/2}$ $18k$ and $3p_{3/2}$ $16k$ Stark levels. At a field of 670 V/cm the lower and upper members of the $3p_{1/2}n=18$ Stark manifold are isolated from the $3p_{3/2}n=16$ Stark manifold, and their configuration interaction is minimal. In this case we observe single peaks corresponding to the $3s$ $18k \rightarrow 3p_{1/2}$ $18k$ transitions, just as was observed in Ba. At 670 V/cm the interior states of the $3p_{1/2}n=18$ Stark manifold are nearly degenerate with states of the $3p_{3/2}n=16$ Stark manifold, and we attribute the observation of multiple peaks to configuration interaction between the two series of Stark states. As shown by Fig. 4 many of the multiple peaks observed are separated by 1.54 cm^{-1}, the spacing between the $n=18$ Stark states;

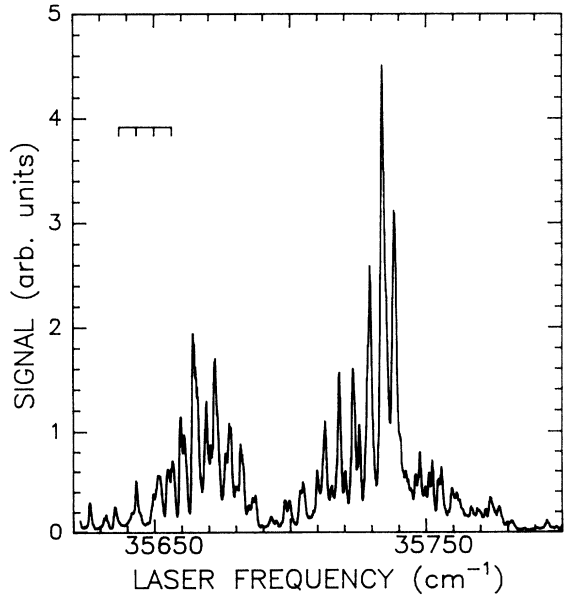

FIG. 3. Spectrum obtained from the interloper in the $n=14$ Stark manifold at a field of 3650 V/cm as a function of the third laser frequency. The two groups of lines are to the Mg $3p_{1/2}$ $14k$ and $3p_{3/2}14k$ Stark states, as can be seen by comparing the observed spacing to the 6.5 cm^{-1} spacing of hydrogenic $n=14$ Stark states shown in the upper left hand corner.

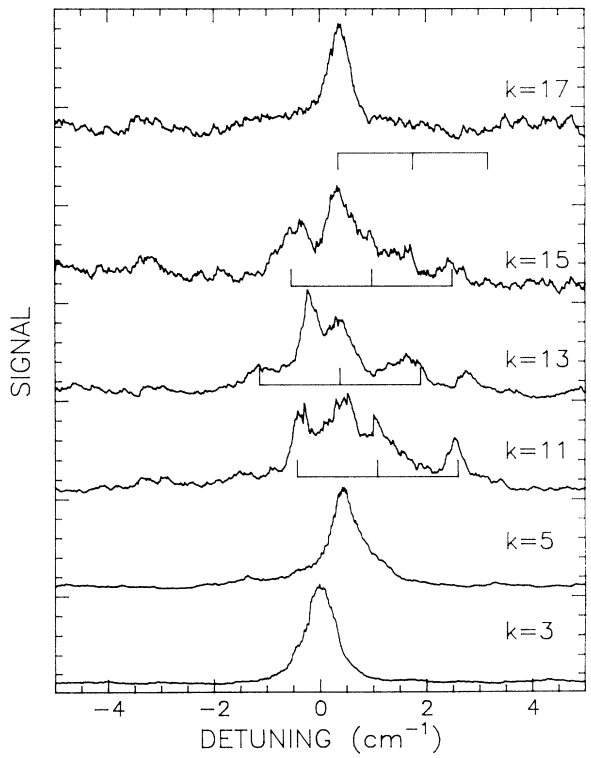

FIG. 4. Observed spectra from the $3s18k-3p_{1/2}18k$ states for $k=3$, 5, 11, 13,15, and 17 in a field of 670 V/cm as a function of the detuning from the Mg^+ $3s_{1/2}-3p_{1/2}$ transition at 35669 cm^{-1}. The $k=3$, 5, and 17 states have spectra consisting of simple Lorentzian lines while the $k=11$, 13, and 15 states have more complex spectra due to the interaction with the $3p_{3/2}$ $16k$ states. Below several of the spectra the $n=16$ and $n=18$ Stark intervals are shown.

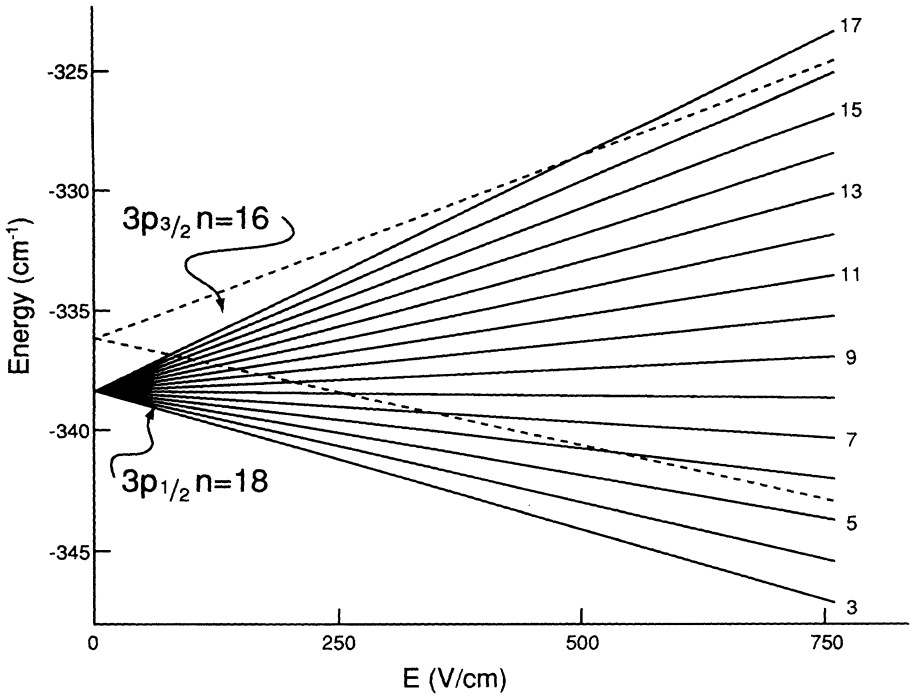

FIG. 5. Energy level diagram of the Mg $3p_{1/2}n=18$ and $3p_{3/2}n=16$ Stark states as a function of electric field. The energies are measured relative to the $3p_{1/2}$ limit, and only the extreme $3p_{3/2}n=16$ levels are shown.

we interpret these as arising from $6p_{1/2}18k$ states. However, it is apparent that there are strong peaks which do not fit into a pattern of lines spaced by 1.54 cm^{-1}. These lines we attribute to $6p_{3/2}16k$ Stark states, which are separated by 1.37 cm^{-1}. These observations suggest that the 18k Stark wavefunction of the initial state has components in several of the $3p_{1/2}n=18$ Stark states due to the configuration interaction with $3p_{3/2}n=16$ Stark state, which also acquires substantial $3p_{1/2}18k$ character. Specifically, we expect that the interaction

$$3p_{1/2}18k \leftrightarrow 3p_{3/2}16k' \leftrightarrow 3p_{1/2}18k'' \quad (1)$$

occurs, leading to the admixture of the original $3p_{1/2}18k$ state into the $3p_{3/2}16k'$ state and other $3p_{1/2}18k''$ Stark states. The admixtures of $18k$ wave function allow the excitation from the initial $3s18k$ state, resulting in multiple peaks in the spectra. The results shown in Fig. 4 are typical. Where the Stark manifolds converging to the Mg^+ $3p_{1/2}$ and $3p_{3/2}$ limits do not overlap we observe single resonances, but where the manifolds overlap we observe multiple peaks.

In general we have found that the widths of the isolated $3snk \rightarrow 3pnk$ resonances, such as those shown in Fig. 4, for $k=3$, 5, and 17 do not vary significantly with the Stark quantum number k, the result obtained previously in Ba. In Ba it was observed that the widths of all the $m=0$ $6p_{1/2}nk$ Stark states were near the average width of the zero-field Ba $6p_{1/2}nl$ states. Furthermore, an approximate n^{-4} scaling was observed for the autoionization widths of the Ba $6pnk$ Stark states [7,8]. No such n scaling is observed in Mg. For example, the $3p_{1/2}$ Stark states have widths of 0.65 cm^{-1} at $n=11$ and 0.45 cm^{-1} at $n=18$, a much slower decrease with n than n^{-4}.

As shown by Fig. 4, many of the spectra taken in static fields exhibit satellite peaks, making the assignment of widths more difficult than it might be and making it difficult to extract an n dependence for the widths. In the hopes of reducing the spectra to ones with only one resonance we have replaced the static field with a 12.8 GHz microwave field with an amplitude two thirds of that required for microwave ionization, i.e., $E_{mw} \simeq \frac{2}{3}(1/3n^5)$ [12]. The field $\frac{1}{3}n^5$ is the field at which Stark states of n and $n+1$ intersect and is approximately the field at which microwave ionization of the bound Mg $3snk$ states occurs. With the microwave field we did in fact observe single resonances, at the ionic transitions, as shown by Fig. 6, even in cases in which a static field of comparable

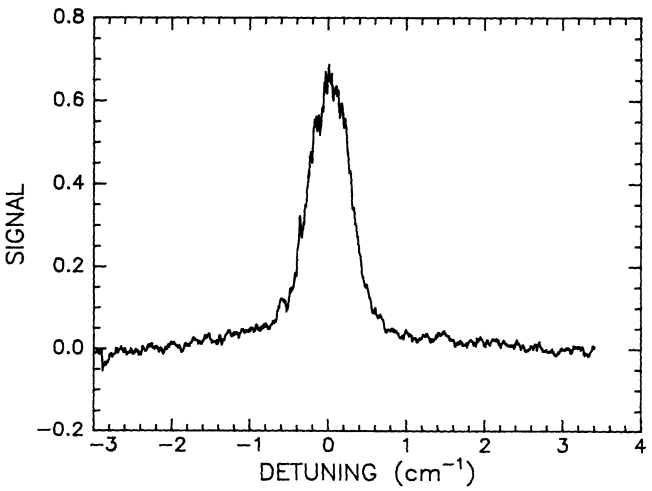

FIG. 6. Observed spectra from the Mg $3s18k$ Stark manifold to the Mg $3p_{3/2}18k$ manifold in a microwave field which is $\frac{2}{3}$ of the microwave ionization field for the bound $n=18$ states of $m=0$ as a function of the detuning from the Mg^+ $3s_{1/2}-3p_{3/2}$ transition at 35761 cm^{-1}.

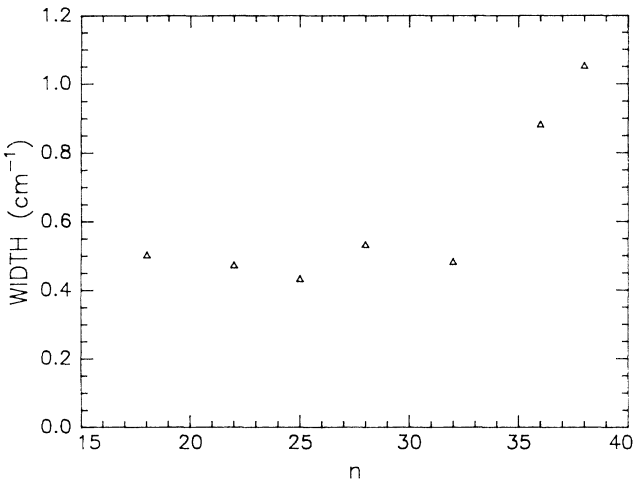

FIG. 7. Observed widths of the Mg $3p_{3/2}$ Stark states in microwave fields of $E \simeq \frac{2}{3}(1/3n^5)$.

size led to multiply peaked structures. From spectra such as those shown in Fig. 6 it is relatively easy to extract widths, and the n dependence of the widths of the $3p_{3/2}$ Stark states is shown in Fig. 7. As shown by Fig. 7, there is almost no dependence of the widths on n, until the $3p_{3/2}$ nk states pass above the Mg^+ $3p_{1/2}$ limit at $n=35$ and have an additional continuum available.

IV. DISCUSSION

The conventional wisdom regarding autoionization of Stark states, which describes the Ba $6pnk$ states quite well, is based on two assumptions. First, the autoionizing Rydberg series are assumed to be converging to one ionic core. The Rydberg nk Stark states are assumed to be composed of linear superpositions of zero-field nl states, and their autoionization rates are the sums of the rates due to their zero-field nl components. Second, the field is assumed not to alter the autoionization rate of the nl state. The contrast between the Mg and Ba results forces us to reexamine the above assumptions. The second assumption seems to be equally valid for Ba and Mg and cannot be the source of the observed difference. The first assumption, however, is not equally good for Ba and Mg. The Δn spacing of the Ba $6pnl$ states of $10<n<20$ is in all cases much less than the 1690 cm^{-1} splitting of the $Ba^+ 6p_{1/2}$ and $6p_{3/2}$ states, but for the Mg $3pnl$ states in the same n range the Δn spacing goes from being greater than to less than the Mg^+ $3p_{1/2}-3p_{3/2}$ interval. Thus Ba is a one-limit problem while Mg is a two-limit problem. In the n regime we have studied the Stark states converging to the Mg^+ $3p_{1/2}$ and $3p_{3/2}$ levels have mixed character and probably have autoionization rates which are averages of the $3p_{1/2}$ and $3p_{3/2}$ rates. Such a mixing of the autoionization rates would lead to a less rapid n dependence of the rates, and we presume this effect to be the dominant reason why the autoionization rates decrease so slowly with n. A second possible contribution to the unexpected n dependence of the widths is that there are excitation amplitudes to both the $3p_{1/2}nk$ and $3p_{3/2}nk$ parts of a Stark state. Since this effect is as likely to lead to increased as decreased apparent widths, we do not think it contributes to the unexpected n dependence of the widths. At high n, greater than 30, we expect that the scalings observed in Ba should also be observed in Mg, since then the Δn interval would be much less than the Mg^+ $3p_{1/2}-3p_{3/2}$ interval. Unfortunately, the separations between Stark states of such high n is far too small for our laser resolution.

V. CONCLUSION

It is evident that the simplest model, based on Rydberg states converging to an isolated ionic core, does not apply in Mg. The autoionization rates in fields do not decrease as rapidly with n as expected. Correspondingly, the dielectronic recombination rate through high-n states should be increased above that predicted by an isolated resonance model. Just as zero-field spectra with substantial interseries interaction have proven to be almost impossible to interpret with an isolated-resonance model [22], we expect that a full analysis of the Mg Stark spectra reported here will require a configuration-interaction approach or a quantum-defect-theory approach [3]. The substantial influence of interseries interaction, even in the presence of a field, suggests that the isolated resonance approximation, commonly used in the calculation of dielectronic recombination rates, may not be adequate and that a more sophisticated approach [23] may be required.

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