Radiation trapping in the far wings of the foreign-gas broadened potassium resonance lines

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We have extended our previous studies of the trapping of alkali-atom resonance radiation to conditions where the unity optical depth points are in the quasistatic wings of the foreign-gas pressure-broadened line shape. Under these conditions, and based on the relatively simple Holstein theory, we expected to see a qualitatively different dependence of the effective radiative decay rate on pressure than is observed when the unity optical depth points are in the impact core. However, we found experimentally that the observed dependence is the same as was observed in the lower pressure limit, at least over the range of conditions we were able to access. On the other hand, these observations are found to be consistent with more accurate calculations based upon the Post theory using a recently available experimental line-shape function.

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INTRODUCTION

We have examined radiation trapping under conditions where the unity optical depth point is in the far wings of the absorption line. In this region far from line center, the quasistatic theory is expected to accurately predict the line shape. High argon pressures ($0<P_{\mathrm{Ar}}<17\,100$ Torr) and potassium densities 3×10^{14} cm$^{-3}<[\mathrm{K}]<1.5\times10^{15}$ cm^{-3}) were used to produce large unity optical depth detunings ($\Delta\nu_{\mathrm{unity}}\sim770$ GHz or $\Delta\lambda_{\mathrm{unity}}\sim15$ Å for the highest argon pressure and potassium density used). Here, the unity optical depth point is defined by $k_\nu l=1$ for $\nu=\nu_0+\Delta\nu_{\mathrm{unity}}$, where k_ν is the absorption coefficient, ν_0 is the line-center frequency, and l is a geometrical factor. These conditions are generally very difficult to achieve in an alkali oven suitable for the study of radiation-trapping effects.

In the experiment described here, we measured the effective fluorescence decay rate of the potassium $4P_{3/2}\rightarrow4S_{1/2}$ resonance transition, following excitation of the upper state by a 0.5-ns laser pulse. The dependence of the decay rates on foreign-gas (argon) pressure was then compared to the predictions of the Holstein [1,2] impact and quasistatic theory expressions and to predictions obtained from the Post theory [3] that were calculated using an experimentally measured line shape (taken from Ref. [4]). We assume that complete frequency redistribution is a valid approximation for the conditions of the experiment (due to the high foreign-gas pressures used).

These new radiation-trapping studies are important, since our experimental conditions more closely approximate than previous studies (Refs. [5–12], for example) conditions found in high-pressure discharge lamps and stellar atmospheres, where trapping is generally dominated by the far wings of the absorption line due to the presence of either large atom densities or long path lengths. Trapping dominated by the quasistatic wings is also of interest because the line profile in this region depends strongly upon the details of the interatomic potentials. Additionally, for conditions involving very high foreign-gas pressures, the trapped decay rates may be affected by nonbinary collisions.

We are aware of only one previous experimental study involving radiation trapping under these high-pressure conditions. In a recent unpublished report by Castex and co-workers [13], radiation trapping (as well as several other processes) was observed in rare-gas mixtures (e.g., Kr-Ar, where krypton was the absorbing atom). In these experiments, synchrotron radiation was used to populate the lower excited levels. For the experimental conditions of those authors, the unity optical depth points lie well beyond the impact region of the line, and thus the trapping is expected to be dominated by the part of the line shape described by the quasistatic theory. Due to the limited pressure range of the experiment, the dependence of the decay rates on buffer gas pressure could be fit equally well with an impact ($[\mathrm{Ar}]^{1/2}$) or a quasistatic ($[\mathrm{Ar}]^{2/3}$) theoretical dependence. (These are the theoretical dependences of the effective radiative rate on buffer gas pressure given by the simple Holstein-theory [1,2] expressions.) The absorbing atom density dependence of the decay rates was found to scale as $[\mathrm{Kr}]^{-1/3}$, consistent with the Holstein quasistatic model. However, the complicated kinetic model needed to describe this system makes it difficult to isolate the effects of trapping from those of other processes. Additionally, the complicated line structure measured by Castex and co-workers [13] for the Kr $^1S_0\rightarrow{}^3P_1$ transition at 1236 Å in the presence of argon buffer gas cannot be handled by the simple Holstein theory. We believe that a more detailed analysis of this system would be required in order to accurately describe the effects of radiation trapping under the conditions of their experiment. We should, of course, note that the study of radiation trapping was not the main focus of the experiments of Ref. [13]. However, it appears to us that the work of Ref. [13] represents the first controlled study of radiation trapping in the quasistatic wings.

EXPERIMENTAL CONDITIONS

Most of the experimental details, including descriptions of the apparatus and the data taking procedures, have been given previously in Refs. [7], [9], and [12]. Further details can be found in Ref. [14].

Alkali vapor (potassium) and buffer gas (argon) were confined in a stainless steel oven with sapphire windows (see Ref. [12]). Two sapphire rods project into the center of the oven (see Fig. 1 of Ref. [12]) to create a slab geometry that approximates Holstein's infinite slab. As in our previous studies, alkali densities have been determined from absorption equivalent width measurements in the present work.

The choice of potassium for the present experiment was determined by the fact that the silver O-ring window seals can withstand a higher density of potassium than sodium. Additionally, both larger broadening rates and slower relative velocities cause the transition from the impact core to the quasistatic wings of the resonance line to occur closer to line center in the potassium case. Thus, for potassium, the trapping is dominated by the quasistatic region of the line at lower argon pressures than in the case of sodium. Finally, the larger fine-structure splitting in potassium simplifies the analysis by avoiding complications due to strong line overlap.

A 0.5-ns pulse from a nitrogen laser pumped tunable dye laser was used to excite the $4\,^2S_{1/2} \to 4\,^2P_{3/2}$ transition in potassium at 7664.91 Å. A 0.5-m monochromator was used to disperse and resolve fluorescence emitted perpendicular to the laser beam. The signal was detected by a photomultiplier with an S-20 spectral response, and time resolution was achieved using a fast transient digitizer (Tektronix model 7912 AD). The effective fluorescence decay rates were obtained from the data by a least-squares fit of a single exponential to each fluorescence signal in the late time following the laser pulse.

High argon pressure and high potassium densities are both required in order to make $\Delta\nu_{\text{unity}} \gg \Delta\nu_c$ so that the quasistatic region of the line dominates the trapping. Here, $\Delta\nu_{\text{unity}}$ is the detuning of the unity optical depth points, where $k_\nu L/2 = 1$, and L is the full slab thickness. The critical frequency detuning $\Delta\nu_c = (2\pi\tau_c)^{-1}$, where τ_c is the duration of a collision, is a measure of the transition between the impact and quasistatic regions of the line [15]. The impact theory is valid in the region where $\Delta\nu \ll \Delta\nu_c$, while the quasistatic theory is valid in the region where $\Delta\nu \gg \Delta\nu_c$. The largest unity optical depth detuning for our experiment was predicted to be $\Delta\nu_{\text{unity}} = 15.7\Delta\nu_c$ (i.e., $\Delta\lambda \sim 44$ Å) for an argon pressure of $\sim$17 065 Torr and a potassium density of 8.9×10^{14} cm^{-3}. We used the Weisskopf theory to determine the average duration of a phase-changing collision, and a quasistatic line shape (where an R^{-6} van der Waals interaction was assumed) for a single pair of atomic interaction potentials to determine the unity optical depth detuning. This unity optical depth detuning is a factor of 1.75 times larger than one calculated using an impact broadened Lorentzian line shape. In either case, broadening rates from Ref. [16] were used.

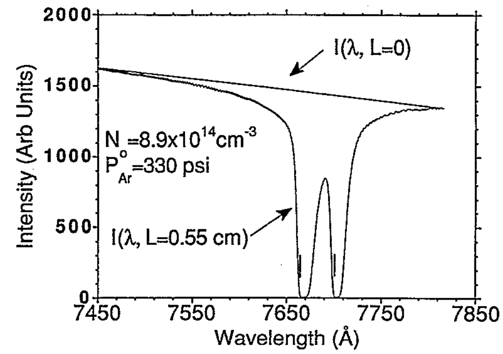

FIG. 1. Absorption equivalent width scan of the potassium $4S_{1/2} \to 4P_{3/2}$ and $4S_{1/2} \to 4P_{1/2}$ resonance transitions for a potassium density of 8.9×10^{14} cm^{-3} and an argon pressure of 17 065 Torr. The path length through the vapor was $L = 0.55$ cm. The absorption equivalent width for the doublet is 60.245 Å in this case. The two vertical marks label the unperturbed line-center wavelengths at 7664.91 and 7698.98 Å.

In fact, the largest observed unity optical depth detuning was $\Delta\nu_{\text{unity}} = 770$ GHz$= 5.4\Delta\nu_c$ ($\Delta\lambda \sim 15$ Å), as seen in Fig. 1. This clearly results from the fact that a pure R^{-6} potential does not fully describe the line shape this far into the line wings. Note that strong absorption occurs over a range of more than 50 Å under these conditions. For these large unity optical depth detunings, the dependence of the trapped decay rates on argon pressure and potassium density was expected to deviate significantly from that predicted for the impact region of the line.

RESULTS AND DISCUSSION

We have compared decay-rate measurements (ω_- is a weighted sum of the trapped radiative decay rates for the two fine-structure levels, as discussed in Refs. [7], [9], and [12]) to predictions of the Holstein impact or quasistatic expressions and to Post-theory calculations based on the experimentally measured line shape from Ref. [4]. We refer to Refs. [7], [9], [12], and [17] for a discussion of our use of these theories of radiation trapping and of our treatment of fine-structure effects. Complete details of the present calculations can be found in Ref. [14]. For conditions of large unity optical depth detunings, described in the present work, we have plotted our measured values of $\omega_-[\mathrm{K}]^{1/2}$ versus argon pressure along with calculated results in Fig. 2. The factor $[\mathrm{K}]^{1/2}$ is included to remove the potassium density dependence from the measured values so that data taken at different densities can be compared. This factor is based upon the Holstein impact theory (valid for the lower pressures of Fig. 2) where decay rates are expected to scale as $[\mathrm{K}]^{-1/2}$. The Holstein quasistatic theory yields decay rates that scale as $[\mathrm{K}]^{-1/3}$. However, the difference between these two dependences cannot be observed over the limited range of potassium densities studied here, given the

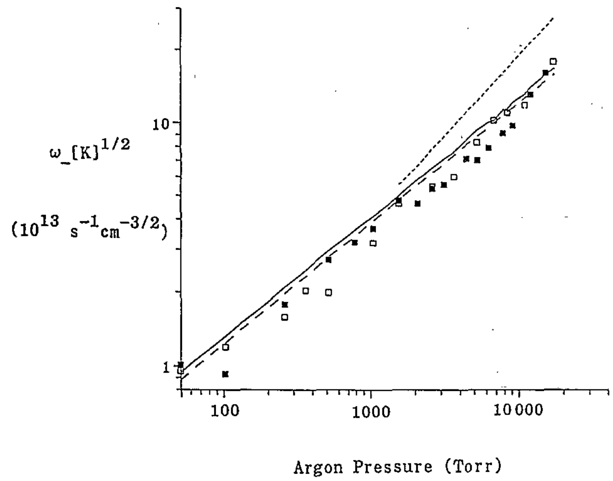

FIG. 2. Results of high argon pressure and high potassium density decay-rate measurements. The quantity $\omega_{-}[K]^{1/2}$ is plotted against the argon pressure. This removes most of the potassium density dependence, which is small in any case, since the data were taken over a small range of densities. The ■ data were taken for an average potassium density of about 5×10^{14} cm^{-3}, and the □ data were taken for an average potassium density of $\sim9\times10^{14}$ cm^{-3}. The solid line represents calculations carried out using the Post theory, as in Ref. [3] (see also Refs. [12] and [14]) but using the line shape from Ref. [4]. The long dashed line is the impact regime Holstein theory as in Ref. [1]. The short dashed line is the quasistatic Holstein calculation (for $[K]=8\times10^{14}$ cm^{-3} and a pure van der Waals interaction). All calculations assume complete frequency redistribution and complete fine-structure level mixing.

present error bars. The difference between the two theories can, however, be clearly seen in the argon pressure dependence (see Fig. 2).

Using arguments similar to those of Ref. [12], we can eliminate quenching by impurities or K_2 molecules as a significant source of uncertainty in the measured decay rates. Quenching of excited potassium by argon atoms is expected to be small, and would lead to decay rates that scale linearly with argon pressure. Instead, as can be seen in Fig. 2, the observed decay rates scale as the square root of the argon pressure. From these data, we can place an upper limit of 7×10^{-20} cm^2 on the cross section for quenching of the potassium $4P$ levels by argon. This value is a factor of 14 lower than the upper limit given in Ref. [18].

With quenching eliminated as a source of uncertainty, we estimate the total uncertainty in our measured decay rates to be $\sim25\%$, due primarily to uncertainty in the potassium density obtained from the equivalent width [14]. The latter, in turn, is uncertain, since it depends on the accuracy of the experimental line shape function [4] under these conditions.

Our preliminary results were difficult to interpret, since the simple quasistatic line shape, considered by Holstein to be based upon van der Waals (or other pure R^{-n}) potentials, did not accurately reproduce our white light absorption measurements. We found that the experimental line shape of Ref. [4] agreed very well with the line shape determined from our equivalent width measurements. Thus we adopted this line shape for use in analyzing our equivalent widths as well as for decay-rate calculations carried out using the more accurate Post theory. It can be seen in Fig. 2 that these more accurate calculations reproduce the measured decay rates quite well over the entire range of conditions studied. The accuracy of the Holstein impact theory has already been well established under conditions equivalent to the lower pressures of Fig. 2. [For the potassium-argon case, it appears that use of slightly smaller broadening rates than those of Ref. [16] would have given slightly better agreement in the present case (see Fig. 2). However, our error bars are such that modification of the broadening rates is clearly unwarranted at the present time.] The continued good agreement between the Holstein-impact-theory results and the experimental decay rates at the highest pressures recorded here is surprising. However, this results from the fact that the experimental line shape is found to be nearly Lorentzian for detunings at least three times larger than the critical detuning $\Delta\nu_c$ (see Fig. 1 and Refs. [4] and [14]). From an extrapolation of the experimental line shape of Kantor, Penkin, and Shabanov [4], it appears that a strong deviation from the Holstein-impact-regime $[Ar]^{1/2}$ dependence would only be observable for argon pressures in excess of 10 000 Torr [14]. Unfortunately, such conditions are only marginally accessible in the present apparatus.

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