Low-pressure Noble Gas Broadening of the Cs Resonance Lines

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The wings of the Cs resonance line ($6\,^2S_{1/2} - 6\,^2P_{1/2,3/2}$) broadened by noble gases (He, Ne, Ar and Kr) at low pressures ($p \leqq 700$ Torr) have been measured in absorption in the range 4 up to a few hundred wavenumbers from the line centres. The far wings are compared with quasistatic wings based on the semiempirical potentials by Baylis and by Pascale and Vandeplanque. In some cases the agreement between the experimental and theoretical line wings is found to be reasonable.

1. Introduction

It is well-known that the shape of the far wings of pressure broadened lines is closely related to the interaction potentials of the colliding atoms through the quasistatic theory [1]. Thus analysis of the spectra can give information on the adiabatic potentials of the upper and lower states. In special cases, if the difference potential is a monotonic function of the interatomic distance, the potentials can even be determined from the spectra by studying the temperature dependence of the far wings. This was shown first by Hedges et al. [2] in an experiment on the broadening of the Cs resonance lines ($6\,^2S_{1/2} - 6\,^2P_{1/2,3/2}$) by noble gases. In the present work we are investigating the same subjects, the broadening of the Cs resonance lines by He, Ne, Ar and Kr. The broadening by Xe has already been studied in our laboratory some years ago [3]. In contrast to Hedges et al. we have measured the spectra in absorption instead of emission. Further, our data are covering not only the far wings but also the transition region from the impact to the statistical wings ($\Delta\bar{\nu} \geqq 0.4\,\mathrm{cm}^{-1}$ from line centre). The shape of the near wings could not be studied by Hedges et al. because of their low instrumental resolution power. In their paper the line wing data are given for $\Delta\bar{\nu} \geqq 70\,\mathrm{cm}^{-1}$ from the line centres. Our main interest is to compare the measured line wings with theoretical data based on interaction potentials by Baylis [4] and by Pascale and Vandeplanque [5] in order to find a set of potentials for a collisional system which can be used for calculations of line wings due to multiperturber effects. The following paper on high-pressure noble gas broadening of the Cs resonance lines will deal with this topic.

2. Experiment

The experimental set-up was simple. The light of a tungsten halogen lamp was transmitted through the absorption cells (length L: 10 cm), containing the Cs vapour-noble gas mixtures, and focussed onto the entrance slit of the Kiel 10 m high resolution Ebert scanning spectrometer (typical band widths: 40 mÅ). The light at the exit slit was detected photoelectrically (photomultiplier: Hamamatsu R 936), the signal amplified and recorded by a strip-chart recorder. By a system of mirrors we could also direct the light of the lamp through a second absorption cell containing only Cs vapour at low pressure. Thus we could determine the positions of the centres of the unperturbed lines.

The sealed-off absorption cells were made out of pyrex glass and were filled with Cs metal and high purity noble gas. For the investigation of each Cs-noble gas system two cells with noble gas pressures of 200 and 700 Torr at room temperature were produced. The cells were placed in a double chamber oven. The temperatures of the chambers could be tuned independently. The cold finger with the Cs metal bath projected into the lower part while the absorption tube was situated in the upper chamber. The temperature in the upper chamber was kept constant at a higher level than the temperatures of the lower chamber which was varied and determined the Cs vapour pressure in the absorption cell. The Cs particle densities were calculated from the vapour pressure curve of Taylor and Langmuir [6]. Additionally the total absorption

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of optically thin higher principal series lines of Cs was measured. By using experimental oscillator strengths [7] and the well-known relation $\int k_\lambda \, d\lambda = (\pi e^2 \lambda^2 f N_{Cs})/m c^2$ the Cs number density could be determined independently. There was always agreement between the densities from the vapour pressure curve and the absorption experiment within 10%. The reduced absorption coefficient $k/N_{Cs} N_{NG}$ of the quasi-static wings are measured by changing the Cs number density in the cells.

The total experimental uncertainty of the measured reduced absorption coefficients is estimated to be of the order of 10–15%. While the statistical errors are about 2–3% and the uncertainty in the noble gas density determination about 1% (measured with a precision manometer), the main uncertainty comes from the Cs density determination (about 10%).

3. The Semiempirical Cs-noble Gas Potentials and the Theoretical Quasistatic Wings

To our knowledge there are no ab-initio calculations of the Cs-noble gas potentials. But there are semiempirical pseudo-potential calculations by Baylis [4], Pascale and Vandeplanque [5] and Czuchaj [8]. All authors – as examples we will only use the sets of data of the two first papers – apply basically the same method. They include the electrostatic interactions as well as the pseudo-interactions [9] of the alkali and noble gas atoms. The one-electron wavefunction of the molecular system is expanded in terms of the alkali valence electron wavefunctions and the potentials are obtained by solving the secular equation. The main difference between the two works cited first is due to the larger number of levels which are included in the basis of Pascale and Vandeplanque compared with Baylis' calculations; the latter took into account only the ground state $6^2S_{1/2}$ and the $6^2P_{1/2,3/2}$ levels. They also used different electronic charge densities of the closed electron cores. The potentials are semiempirical because their evaluation requires the experimental values of various parameters. These parameters are the energy levels of the alkali atom and the polarisabilities of the noble gas atoms which can be obtained from spectroscopic studies and semiempirical calculations, respectively. The third parameter is a measure of the size of the noble gas atoms which can be taken from alkali-noble gas scattering data.

The Cs-noble gas potentials $X^2\Sigma_{1/2}$, $A^2\Pi_{1/2}$, $A^2\Pi_{3/2}$ and $B^2\Sigma_{1/2}$ as derived by Baylis [4] and by Pascale and Vandeplanque [5] are displayed as full (Baylis) and dashed curves (Pascale and Vandeplanque) in the left parts of Figure 1–4. One recognizes that there are significant differences in the potentials of Baylis and of Pascale and Vandeplanque. First, the potentials of Pascale and Vandeplanque are less repulsive compared with those of Baylis, with the exception of the Cs-Ne system. This fact reflects the larger sizes of the electronic density distributions chosen by Baylis. Second, the larger basis set used by Pascale and Vandeplanque strongly modifies the shapes of the potentials. In particular, the couplings with the nearby higher-lying states forces the $B^2\Sigma_{1/2}$ potentials down. But

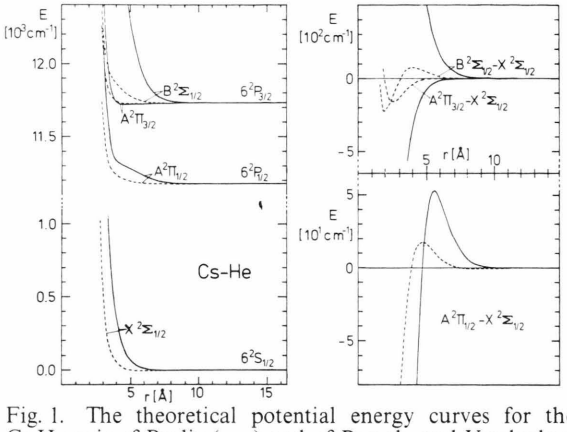

Fig. 1. The theoretical potential energy curves for the Cs-He pair of Baylis (——) and of Pascale and Vandeplanque (– – –), and the relevant difference potential curves.

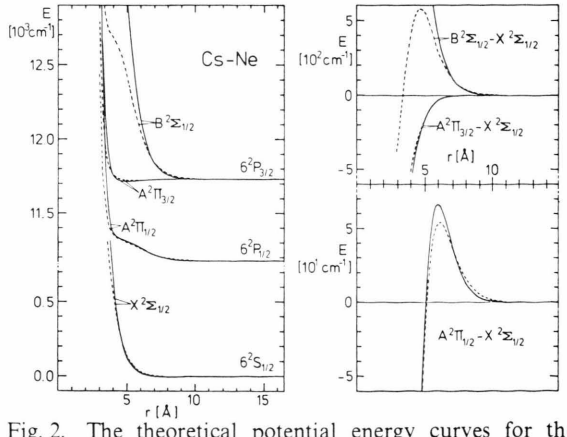

Fig. 2. The theoretical potential energy curves for the Cs-Ne pair. Baylis (——) and Pascale and Vandeplanque (– – –).

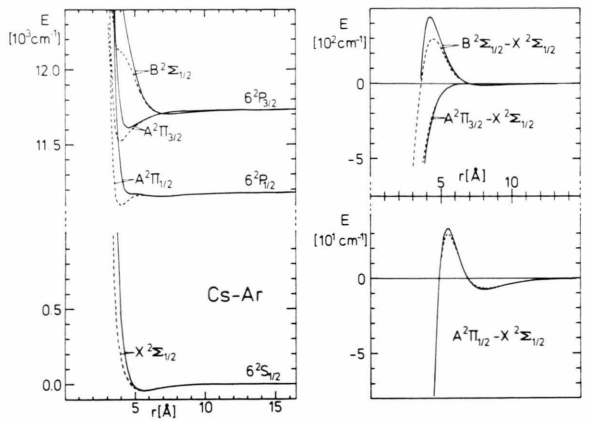

Fig. 3. The theoretical potential energy curves for the Cs-Ar pair (—, Baylis and – – –, Pascale and Vandeplanque).

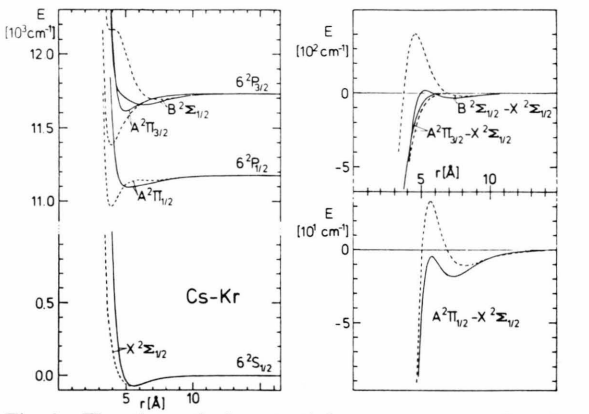

Fig. 4. The theoretical potential energy curves for the Cs-Kr pair. (—, Baylis and – – –, Pascale and Vandeplanque).

the effect of coupling cannot be neglected for the other potentials either.

On the right sides of Fig. 1–4 we have plotted the differences of the upper $V_u(r)$ and lower potentials $V_l(r)$ with respect to the energy differences of the $6^2P_{1/2,3/2}$ and $6^2S_{1/2}$ levels against the interatomic distance r:

$$h\,c\,\Delta\bar{\nu} = V(r) = V_u(r) - V_l(r)\,. \tag{1}$$

These difference potentials $V(r)$ can be used to calculate the quasistatic wings of the noble gas broadened Cs resonance lines.

In the quasistatic theory the absorption coefficient $k(\bar{\nu})$ in a small energy interval $(\bar{\nu}, \bar{\nu}+\Delta\bar{\nu})$ of the line wing is connected via the interatomic separation r to the probability $W(r)\,\mathrm{d}r$ of finding the nearest noble gas perturber in a differential part $\mathrm{d}r$ at the interatomic distance r:

$$k(\bar{\nu})\,\mathrm{d}\bar{\nu} \sim W(r)\,\mathrm{d}r\,. \tag{2}$$

The absorption coefficient in the line wing is given by

$$k(\bar{\nu}) \sim h\,c\,W(r)/|\,\mathrm{d}V(r)/\mathrm{d}r\,|\,. \tag{3}$$

inserting formula (1) into relation (2).

For $W(r)\,\mathrm{d}r$ we choose the simple binary approximation, including the Boltzmann-factor of the ground state potential

$$W(r)\,\mathrm{d}r = 4\pi r^2 N_{NG} \cdot \exp\left(-4\pi r^3 N_{NG}/3 - \Delta V_l(r)/kT\right)\mathrm{d}r\,. \tag{4}$$

To a good approximation $\exp(-4\pi r^3 N_{NG}/3)$ is unity in the relevant interatomic range, taking into account our experimental noble gas densities N_{NG}. The Boltzmann factor $\exp(-\Delta V_l(r)/kT)$ can be simply used because the potential depths of the Cs-noble gas ground state potentials are very shallow (dissociation energy $< kT$). Therefore we have not to discriminate between bound and free particle distributions [2]. Together with the normalisation of the absorption line

$$\int k(\bar{\nu})\,\mathrm{d}\bar{\nu} = \frac{\pi e^2}{m c^2} N_{Cs} f \tag{5}$$

we get the reduced absorption coefficient $k(\bar{\nu})/N_{Cs}N_{NG}$ (in $\mathrm{cm}^{-1}/\mathrm{cm}^{-6}$) in the line wing

$$k(\bar{\nu})/N_{Cs}N_{NG} = \frac{4\pi^2 r^2 e^2 h f}{m c} \frac{\exp(-\Delta V_l(r)/kT)}{|\,\mathrm{d}V(r)/\mathrm{d}r\,|}\,. \tag{6}$$

If more than two difference potentials are responsible for the shape of the wings (example: the $6^2S_{1/2} - 6^2P_{3/2}$ transition) we have to add both contributions taking into account the statistical weights of the molecular states. In our simple classical approximation we make some assumptions:

(i) The oscillator strength f is assumed to be independent of the interatomic distance, which is not strictly true.

(ii) The condition $N_{Cs} \ll N_{NG}$ has to be fulfilled. In the foreign gas broadening experiment one has to make sure that the absorbing atoms interacting only with perturber atoms and not with atoms of the same species. If the condition $N_{Cs} \ll N_{NG}$ is not satisfied self-broadening effects which are significant for the resonance lines have to be taken into account.

(iii) The absorbing atom interacts only with one perturber atom at a time because we like to compare the measured with calculated quasistatic wings based on binary interaction potentials. Otherwise multiperturber interactions (the topic of the subsequent paper) have to be taken into account.

Further, because we do not allow for thermal motion of the interacting particles, non-adiabatic transitions to nearby states are neglected during the collision. In the far and very far line wings where we get information about the potentials at small interatomic separations these mixing collisions can be neglected in most cases.

The appropriate theoretical description of the line wings would be given if we would use the quantum mechanical, unified Franck-Condon treatment of Szudy and Baylis [10]. In particular, in the regions of satellites, where the difference potentials $V(r)$ have extrema and $k(\bar{\nu}) \to \infty$ (see formula (6)), realistic line shapes can be calculated using the unified theory. The sharp singularities of the quasistatic approximation disappear. The satellites are smeared out and the exponential fall-off of the absorption coefficient beyond the satellites, a classical forbidden region, is also properly given. However, if there are no singularities in the spectrum, the line shapes of the quasistatic approximation and the unified theory differ little. As mentioned above, we are more interested in a check of the theoretical difference potentials rather than in describing the satellite shapes in an appropriate way. Therefore the comparison between our experimental profiles and the theoretical quasistatic wings will give sufficient information.

4. Results and Discussion

In Figs. 5–12 the measured reduced absorption coefficients $k/N_{Cs}N_{NG}$ of the wings of the 8944 Å ($6^2S_{1/2} - 6^2P_{1/2}$) and 8521 Å lines ($6^2S_{1/2} - 6^2P_{3/2}$) are plotted against the distance $\Delta\bar{\nu}$ (in cm^{-1}) from the unperturbed line centres. The data were taken at different Cs particle densities under optically thin conditions. The dots and the triangles correspond to data determined at noble gas pressures of 200 and 700 Torr, respectively. The quasistatic line wings based on the Cs-noble gas potentials by Baylis [4] and by Pascale and Vandeplanque [5] and calculated using formula (6) are shown as full (Baylis) and dashed line shapes (Pascale and Vandeplanque) in Figures 5–12. The oscillator strengths f were taken from [11]. For comparison we give also the experimental data of Hedges et al. [2] at $-192\,cm^{-1}$ (8944 Å line) and $-320\,cm^{-1}$ (8521 Å line) converted to our experimental conditions ($T = 503$ K). Taking into account the mutual error bars, there is

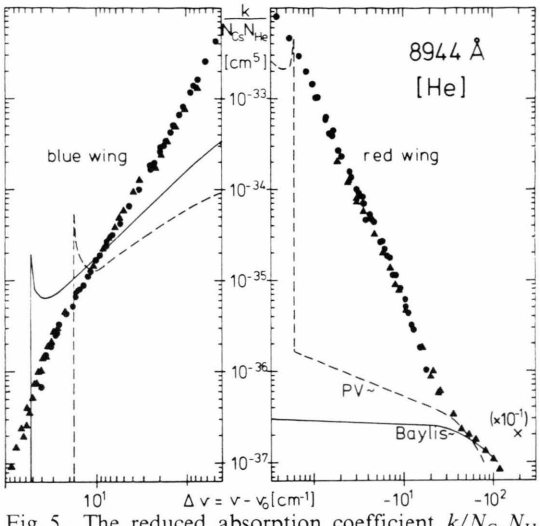

Fig. 5. The reduced absorption coefficient $k/N_{Cs}N_{He}$ of the wings of the 8944 Å line broadened by He. The data were taken at He gas pressures of 200 (●) and 700 Torr (▲). The quasistatic wings based on the semiempirical potentials of Baylis (——) and of Pascale and Vandeplanque (– – –) are given for comparison. The value (X) at $\Delta\bar{\nu} = -192\,cm^{-1}$ is taken from [2].

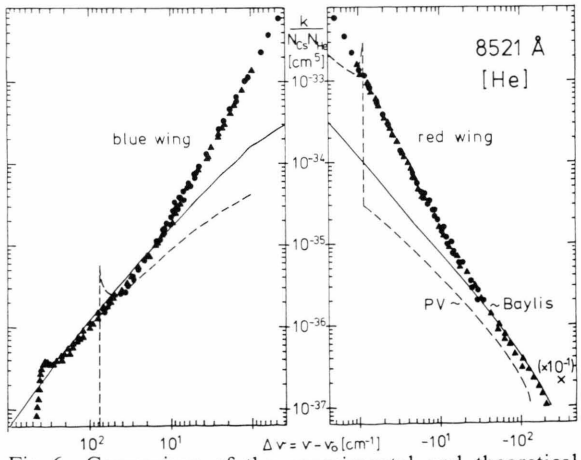

Fig. 6. Comparison of the experimental and theoretical wings for the 8521 Å line broadened by He. See Fig. 5 for comments.

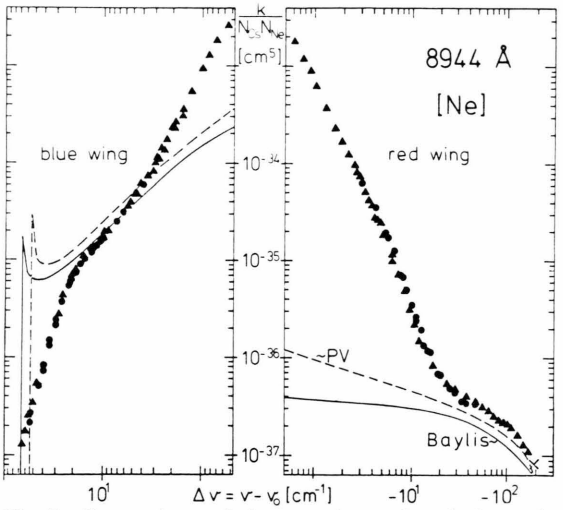

Fig. 7. Comparison of the experimental and theoretical wings for the 8944 Å line broadened by Ne. See Fig. 5 for comments.

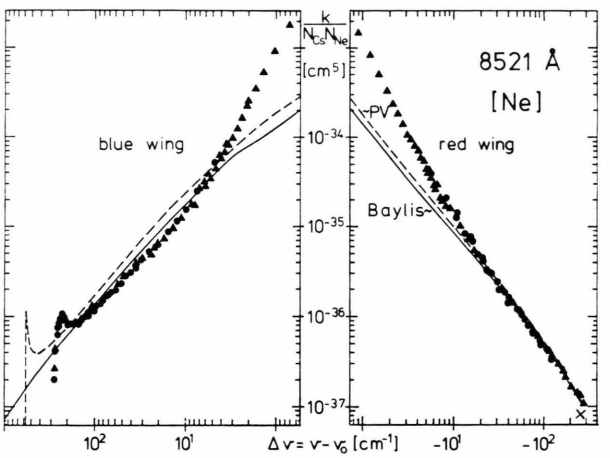

Fig. 8. Comparison of the experimental and theoretical wings for the 8521 Å line broadened by Ne. See Fig. 5 for comments.

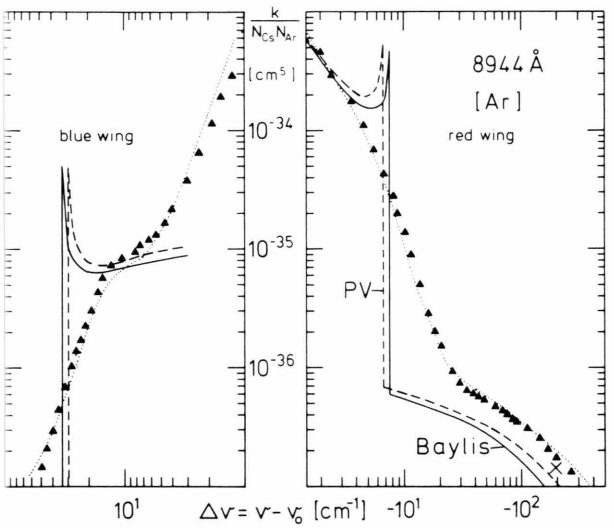

Fig. 9. Comparison of the experimental and theoretical wings for the 8944 Å line broadened by Ar. (· · ·) are experimental data of Chen and Phelps [14]. See Fig. 5 for comments.

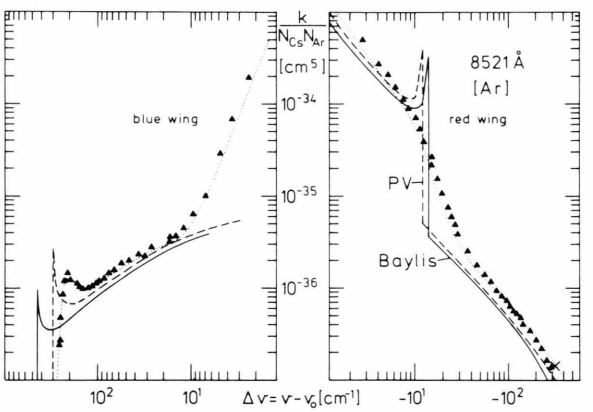

Fig. 10. Comparison of the experimental and theoretical wings for the 8521 Å line broadened by Ar. (· · ·) are experimental data of Chen and Phelps [14]. See Fig. 5 for comments.

agreement between the data for the systems Cs-Ne, Cs-Ar and Cs-Kr. In the case of Cs-He we did not measure so far in the wings.

How the shapes of the lines change with $\Delta\bar{\nu}$ can be seen in Figures 5–12. Near to the line centres the wings are Lorentzian in shape ($k/N_{\mathrm{Cs}}N_{\mathrm{NG}} \sim \Delta\bar{\nu}^{-2}$). Small asymmetries which can hardly be seen in the figures are mainly due to the impact shift of the lines with respect to the unperturbed line positions. It is interesting to calculate the impact broadening widths γ/N_{NG} from the measured data in the Lorentzian region by (see e.g. [12])

$$\gamma/N_{\mathrm{NG}} = \frac{2m\,c^2\,(\Delta\bar{\nu})^2}{e^2 f}\,\frac{k}{N_{\mathrm{Cs}}N_{\mathrm{NG}}} \tag{7}$$

and compare the data with values of other authors. In Table 1 our γ/N_{NG} data are listed together with

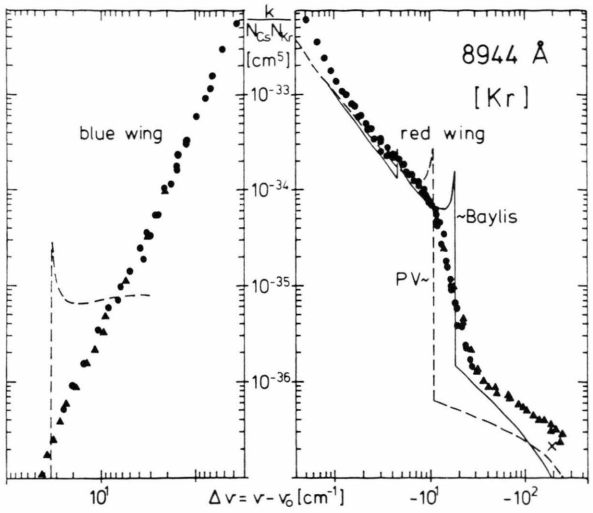

Fig. 11. Comparison of the experimental and theoretical wings for the 8944 Å line broadened by Kr. See Fig. 5 for comments.

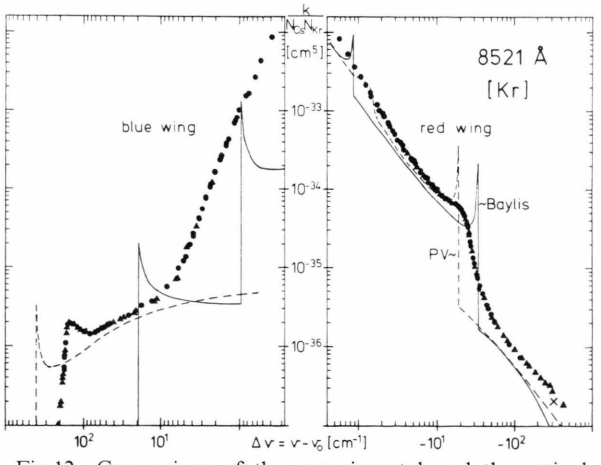

Fig. 12. Comparison of the experimental and theoretical wings for the 8521 Å line broadened by Kr. See Fig. 5 for comments.

Table 1. A comparison of our broadening rates γ/N (10^{-20} cm^{-1}/cm^{-3}) with the data of Bernabeu and Alvarez [13]. Our experimental temperature was about $T = 390(20)$ K (Bernabeu and Alvarez: $T = 295$ K).

	$6\,^2S_{1/2} - 6\,^2P_{1/2}$		$6\,^2S_{1/2} - 6\,^2P_{3/2}$	
	This work	Ref. [13]	This work	Ref. [13]
He	1.04(16)	0.98(7)	1.21(18)	1.36(9)
Ne	0.63(10)	0.51(4)	0.52(8)	0.53(11)
Ar	1.14(17)	1.00(11)	0.91(25)	1.16(1)
Kr	1.04(16)	1.01(12)	0.55(8)	0.55(5)

more recent data by Bernabeu and Alvarez [13]. The data agree within their mutual error bars. The transition regions from the Lorentzian cores to the statistical wings can be taken from the spectra (Figs. 5–12). They were found to be about 10, 4, 2 and 1 cm^{-1} for the perturber He, Ne, Ar and Kr, respectively.

For the system Cs-He (Figs. 5 and 6) all features observed in the wings of the 8944 Å line can also be seen in the theoretical wings. There is a broad, smeared-out satellite in the blue wing due to an extremum in the $A\,^2\Pi_{1/2} - X\,^2\Sigma_{1/2}$ difference potential (see Fig. 1). Note that the position of the satellite is not exactly given by the theoretical wings. Further, there is a rapid fall-off beyond the Lorentzian region in the red wing due to nonstatic effects and the far red statistical wing which is excellently described by the quasistatic wings including Baylis' difference potential. Comparing the wings of the 8521 Å line with the theoretical wings (Fig. 6) we recognize that the blue satellite is predicted by Pascale and Vandeplanque but the theoretical position deviates from the found position by about 300 cm^{-1}. Because Baylis did not take into account higher levels in his calculation, his difference potential $B\,^2\Sigma_{1/2} - X\,^2\Sigma_{1/2}$ does not show an extremum (Fig. 1) and therefore no satellite should be seen in the spectrum. The satellite in the quasistatic red wing of Pascale and Vandeplanque is located in the impact region and cannot be detected. The shape of the far red wing is qualitatively given by the theoretical wings.

Comparing the experimental wings of the Cs resonance lines broadened by Ne (Figs. 7 and 8) with the theoretical wings the situation is similar to that in Cs-He. There is again a satellite in the blue wing of the 8944 Å line which is predicted by the theory. But as in the case of Cs-He the expected position of the satellite could not be verified by the experiment. On the other hand, the shape of the far red wing is predicted by theory. The red wing of the 8521 Å line (Fig. 8) is in excellent agreement with both theoretical wings. As for Cs-He, the blue wing is qualitatively well described by the quasistatic wings based on Pascale and Vandeplanque's potentials. Again there is a disagreement in the positions of the satellite and there is no satellite in the quasistatic wings calculated from Baylis' difference potentials. We note that the blue satellite arises from transitions at an interatomic separation of about 4.5 Å

(see Figure 2). Here the height of the extremum in the $B^2\Sigma_{1/2} - X^2\Sigma_{1/2}$ difference potential curve depends in a very sensitive way on the slopes of the repulsive $X^2\Sigma_{1/2}$ and $B^2\Sigma_{1/2}$ potentials.

There is satisfactory agreement comparing the wings of the Cs resonance lines broadened by Ar – which have also been measured by Chen and Phelps (dotted wings) – with the theoretical quasistatic wings of Baylis and of Pascale and Vandeplanque (Figs. 9 and 10). The shapes of the wings including the approximate positions of the satellites are predicted by theory. The differences in the reduced absorption coefficients are small. As expected from the similar course of the difference potentials of Baylis and of Pascale and Vandeplanque (Fig. 3) there are only small differences in the theoretical quasistatic wings. But note that the Cs-Ar potentials of Baylis and of Pascale and Vandeplanque have completely different forms (Figure 3). Although predicted by Pascale and Vandeplanque, no satellite was found in the blue wing of the Kr broadened 8944 Å line (Figure 11). Beyond the impact region the reduced absorption coefficient of the blue wing varies approximately as $(\Delta\bar{\nu})^{-2.3}$ which is more in line with the dependence for the nonstatic regions predicted by Lindholm [15] than by Tvorogov and Fomin [16]. The satellite in the red wing of the line was expected, taking into account the theoretical potentials. Its position is in accordance with the position of the quasistatic satellite of Pascale and Vandeplanque. Both theories do not give satisfactory data for the far red wing $\Delta\bar{\nu} > |-30|\ \mathrm{cm}^{-1}$. Whether there is a red satellite at $-4.6\ \mathrm{cm}^{-1}$, due to an extremum in the $A^2\Pi_{1/2} - X^2\Sigma_{1/2}$ difference potential curve of Baylis, cannot be decided from the experimental data. The wings of the 8521 Å line broadened by Kr (Fig. 12) are qualitatively well described by the quasistatic wings calculated from the potentials of Pascale and Vandeplanque. But again the absolute data and the positions of the blue and red satellites are not in agreement. Comparing the experimental with the theoretical wings based on Baylis' potentials we see that the first theoretical blue and red satellites at 0.95 and $-0.85\ \mathrm{cm}^{-1}$ are located in the impact core of the line and therefore they are not expected to be seen. While the theoretical red wing (Baylis) is in qualitative agreement with experiment, the blue wing, including the position of the satellite, disagree strongly from the observed shape.

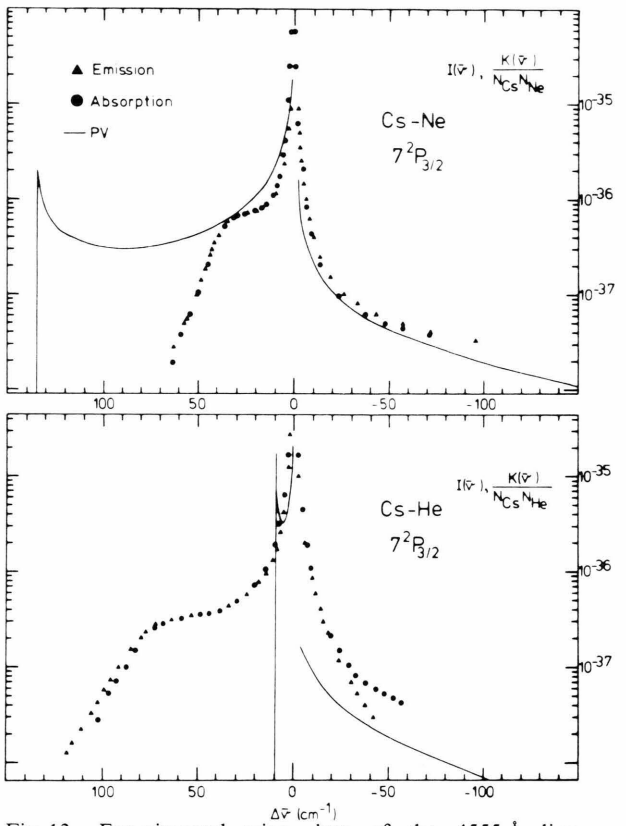

Fig. 13. Experimental wing data of the 4555 Å line ($6^2S_{1/2} - 7^2P_{3/2}$) broadened by He and Ne. The data were obtained from absorption (●) and emission measurements (▲). The quasistatic wings based on the potentials by Pascale and Vandeplanque (PV) are shown as full curves.

5. The Noble Gas Broadening of Higher Members of the Principal Series Lines of Cs

By comparing experimental with theoretical wings it was already shown before (Ref. [3]: Cs-Xe) that the quasistatic wings of higher Cs principal series lines based on the potentials by Pascale and Vandeplanque are of poor quality. In addition to the resonance lines we have also investigated the noble gas broadening (He, Ne, Ar and Kr) of the second doublet of the series, $6^2S_{1/2} - 7^2P_J$. The findings of the earlier Cs-Xe experiment were confirmed. On the average, the shapes of the theoretical wings deviate stronger from the found behaviour than in the case of the resonance lines. Only for the lighter noble gases He and Ne are the experimental and theoretical wings still in qualitative agreement. For example, in Fig. 13 we show the comparison of the wings of the 4555 Å line ($6^2S_{1/2} - 7^2P_{3/2}$) broadened

by He and Ne. The experimental data were taken in absorption (●) as well as in emission (▲). The emission experiment was performed by shining the light of the 4579 Å line of an Ar^+-laser into the cell containing the Cs-noble gas mixture. The collisionally redistributed light from the 7P-6S transition was recorded photoelectrically by scanning a monochromator across the interesting wavelength region. The absorption and emission data are in good agreement. Only in the far wings deviations could be detected. The deviations could be due to different Boltzmann factors. For both noble gases the theory predicts blue satellites in agreement with observation. However the predicted positions are far from the real positions.

6. Conclusion

This work presents another example of how the simple quasistatic theory can check the quality of theoretical interaction potentials (better: difference potentials) by comparing the theoretical wings with experimental shapes of pressure broadened lines. In the present paper we show that the semiempirical Cs-noble gas potentials of Baylis and by Pascale and Vandeplanque describe qualitatively the observed broadening of the Cs resonance lines. Generally, the quasistatic wings based on the more advanced potentials of Pascale and Vandeplanque are in better agreement with experiment than those with Baylis' potentials. In some cases even the absolute data of the theoretical reduced absorption coefficients were found to be in good agreement with experiment, except for the region of the position of the satellites. For higher lines of the principal series of Cs the semiempirical potentials (Pascale and Vandeplanque) are of poorer quality. Ab initio calculations of the interaction potentials are badly needed.

We acknowledge gratefully financial support by the Deutsche Forschungsgemeinschaft.

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