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Broadening and shifts of the xenon line at $\lambda = 3.51$ μm: Measurements and theoretical interpretation

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Broadening and shifts of the Xe I line at $\lambda = 3.51$ μm are measured at two temperatures and for various perturbers, He, Ne, Ar, and Xe, by use of a laser linear-absorption experiment. A theoretical interpretation is proposed which involves the quantum theory of collisions. The long-range part of the interaction potential is described by the successive orders of the multipole expansion of the interaction between excited-state atoms and ground-state perturbers; the short-range part is described by an extension of a Fermi-type potential, and a cutoff function is used for the intermediate case. For all perturbers, calculated broadenings and shifts are shown to be sensitive to the position of the potential well, but not to its depth.

I. INTRODUCTION

Broadening and shift of spectral lines, which have been calculated by use of collision theory so far, mainly concern simple-spectra elements in the presence of inert gases since, in these cases, the interaction between colliding partners can be described *ab initio* with some reliability: alkalis in the presence of noble gases, for instance.[1,2] The situation is much more complex in the case of the interaction between noble gases. First, more than one optical electron is involved and second, dipole transitions generally occur between excited states whose coupling scheme is not simply described. However, their study is of interest also, at least from an experimental viewpoint, since it is possible to vary independently several physical parameters which characterize the collision, such as temperature. To interpret the measurements, simple but not realistic interaction potentials have been tried in the past (van der Waals, Lennard-Jones). In a limited number of cases, *ab initio* potentials have been used also, for low-excited states only.[3,4] It is thus of interest to try potential models which are based on the simplification due to the different sizes of the colliding partners.

For the Xe I line at $\lambda = 3.51$ μm $[5p^5 5d(7/2)_3 \rightarrow 5p^5 6p(3/2)_2]$, which is considered in this paper, the measurement of several broadening constants was published in a previous paper which presented also the experimental technique of recording and analysis.[5] Here we present a complete set of results concerning broadenings and shifts of this line. For two perturbers (He and Ar) measurements have been extended to two temperatures. Introduction of simple interaction potentials into classical impact theories is not enough to interpret the data. Therefore, we have tried more realistic potentials, where the long-range interaction is described by the multipole expansion of the excited-state atoms, the short-range interaction by an extension of a Fermi-type potential, and a cutoff function is introduced for the intermediate case.[6] Then the quantum theory of collisions has been used to calculate broadenings and shifts of the line. Also, position and depth of the potential well have been parametrized to observe their influence on calculations.

II. EXPERIMENTAL RESULTS

The experiment which has been used for these measurements has been described in a previous publication.[7] It consists mainly in using a single-mode xenon laser oscillating at $\lambda = 3.51$ μm and finally tunable across its gain curve. The laser beam propagates inside a discharge tube filled with ^{136}Xe and weakly excited by a square-wave current. A lock-in detection is used to record the amplified part of the laser flux only, hence the line profile is obtained when the laser frequency is swept. Fabry-Perot fringes provided by a multipath spherical etalon are simultaneously recorded to calibrate the frequency scale. With this technique one records the total amplification

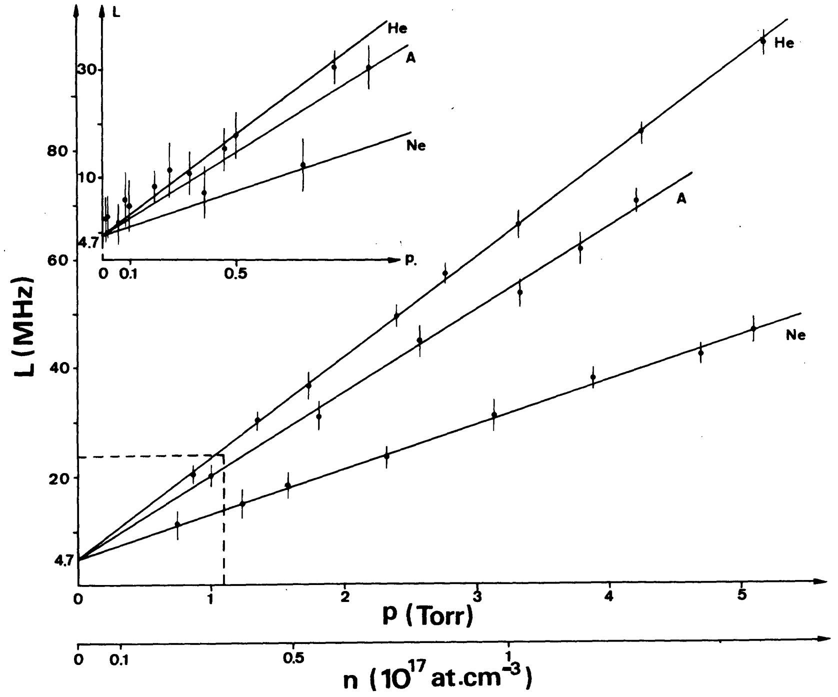

FIG. 1. Variations of the Lorentzian widths with pressure and density of perturbers (He, Ne, Ar). Units of ordinates are in MHz, units of abscissas are in Torr and 10^{17} atom/cm^3. In the insert, the low-pressure domain is represented. Results were obtained at room temperature.

profile of the line, i.e., a Voigt profile, convolution of the homogeneous profile by the Doppler contour. After careful analysis of the data,[5] one deduces the width of the two components, a Gaussian width related to the Doppler effect and a Lorentzian width related to the natural profile broadened by the effect of phase-interrupting collisions; hence, the broadening constants when the pressure of perturbers is varied. Pressure shifts are measured by adding to the setup a second discharge tube which is crossed by another laser beam provided by reflection on a beam splitter. By varying the perturber pressure inside the second tube, while it is maintained constant inside the first one, one is able to deduce shift constants by simply comparing the relative position of the two recorded profiles. This is made directly by help of the Fabry-Perot fringes. All measurements have been performed at room temperature for He, Ne, Ar, and Xe perturbers and at liquid-nitrogen temperature for He and Ar. It has not been possible to evaluate the influence of Kr because nonresonant excitation transfers alter the Doppler profile which no longer remains Gaussian,[8,9] and make it impossible to perform correctly the profile analysis. The densities of perturbers which have been used lie typically in the range 10^{15}–10^{17} atoms/cm^3.

Broadening constants are directly deduced from the variations with pressure of the Lorentzian widths as shown in Fig. 1, for He, Ne, and Ar perturbers at room temperature.[5] In the case of Xe, a perturbation of the Doppler component at low pressure leads to a weak deviation from the linear variation of the Lorentzian (Fig. 2) but the determination of the broadening constant is still possible. For all perturbers, extrapolation to zero density leads to a natural width (4.7 ± 0.5 MHz) in good agreement with the value (4.6 MHz) expected from lifetime measurements[10] and calculations.[11] At liquid-nitrogen temperature, the profile analysis leads to linear variations of Lorentzian widths with pressure and to regular increases of the Gaussian widths, both for He and for Ar (Fig. 3). In the case of He, for instance, the temperature is raised from 80 to 105 K between zero and 4 Torr. The variation is even more rapid in the case of Ar. These temperature variations with pressure can be attributed to the whole discharge or to active atoms only (Xe). In the first case, the variations of the Lorentzian widths become nonlinear with density, even when one takes into account the variation of broadening constants with temperature. Consequently, this assumption must be ruled out. In the second case, the temperature relative only

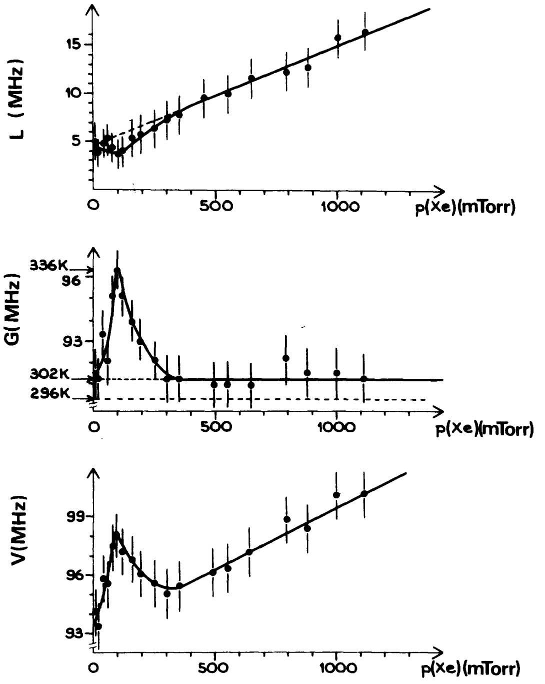

FIG. 2. Variations of the Lorentzian width (L), Gaussian width (G), and width of the Voigt profile with pressure of xenon. Units of ordinates are in MHz, units of abscissas in mTorr. The increase of the Doppler width at low pressure corresponds to a change of temperature of 34 K.

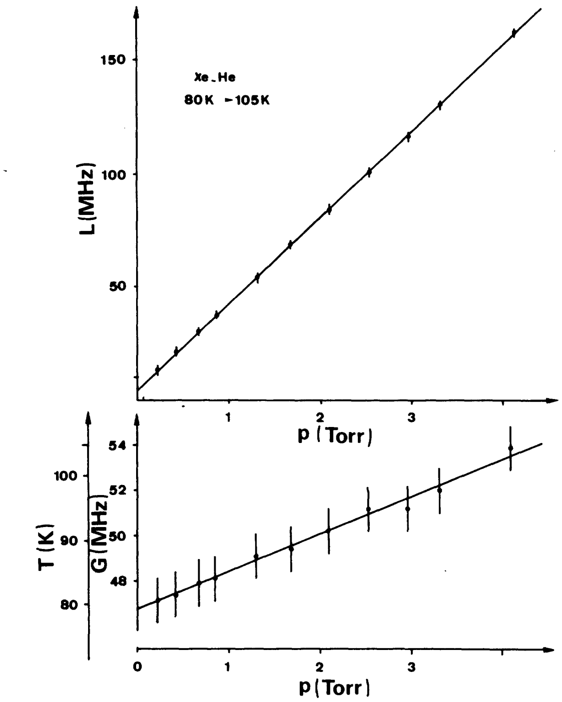

FIG. 3. Variations of the Lorentzian width (L) and Gaussian width (G) with pressure of He, at liquid-nitrogen temperature. Units of ordinates are in MHz, units of abscissas are in Torr. The vertical scale on the left of the lower graph represents the temperature corresponding to the observed Doppler width.

to the Xe active atoms should be modified with pressure, the temperature relative to the bath being constant. Thus the variations of the Lorentzians are again linear functions of perturber densities. The origin of the perturbation could be found in an increase of the electronic temperature, electronic collisions being mainly responsible for the population inversion in xenon discharges. With our experiment, we have no means, however, to verify this assumption but we would like to point out that the phenomenon has been observed also in pure helium where it is even more pronounced.[12]

Shift measurements have been performed under the same experimental conditions. In Fig. 4, the variations of the shifts with perturber densities are plotted. One also notices here that, actually, at liquid-nitrogen temperature, shifts are proportional to pressure and are proportional to density only in the case where active atoms are heated by some mechanism. Shifts are toward high frequencies when increasing the pressure, for all perturbers except He, as already observed in xenon[13-15] but in opposition to the sense which is generally observed.[16]

Results of broadening and shift measurements are summarized in Table I, in terms of γ/n and d/n, respectively, where n is the density of perturbers. T is the temperature of the discharge, deduced from measurements of the Doppler width. The sign "−" for shifts indicates shifts towards low frequencies ("red" shifts).

III. USE OF CLASSICAL IMPACT THEORIES TO INTERPRET THE DATA

Measurements have been performed in a range of perturber densities where typically the impact approximation holds very well. We are allowed to consider also, that inelastic collisions if they exist, are very rare compared to elastic collisions.[17] Thus, we have tried in a first step to interpret the data by use of the classical Lindholm-Foley impact theory of broadening[18] where we have introduced a simply attractive van der Waals interaction potential ($-C_6/R^6$). In this case, broadening constants can be expressed as

$$\gamma/n = 0.795\times 10^5(\bar{v})^{3/5}|\Delta C_6|^{2/5}, \qquad (1)$$

where γ is in MHz, n in atoms per cm^3; $\bar{v}$ is the

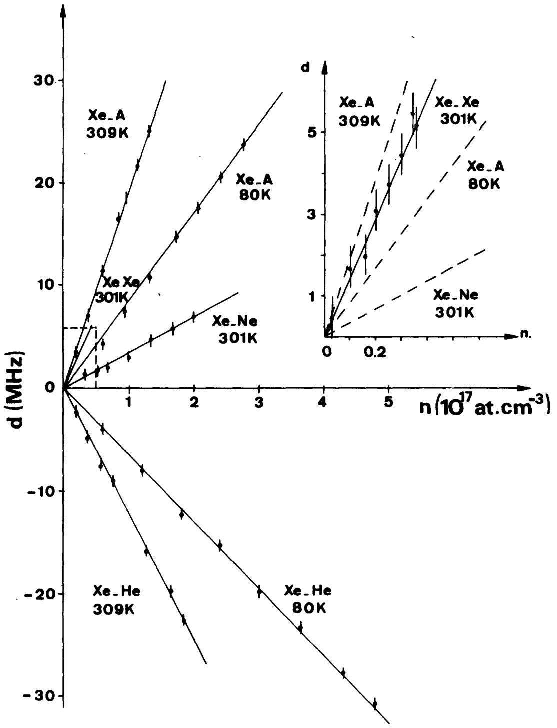

FIG. 4. Pressure shifts measured as functions of perturber density. Units of ordinates are in MHz, units of abscissas are in 10^{17} atom/cm^3. In the insert, the low-pressure domain is represented with Xe measurements, in particular.

mean relative velocity between colliding partners, in cm per s, and ΔC_6 is the van der Waals constant related to the transition, in erg cm^6. ΔC_6 can be estimated from

$$\Delta C_6 = e^2\alpha\left(\sum\langle r_f^2\rangle - \sum\langle r_i^2\rangle\right), \qquad (2)$$

e being the electronic charge, α the dipole polarizability of perturbers in their ground state, and $\langle r^2\rangle$ the mean-square radius of optical electrons in final and initial states. The sum over $\langle r^2\rangle$ is extended to all optical electrons. α and $\langle r^2\rangle$ values can be found in Dalgarno's tables[19] and Aymar's calculations,[20] respectively.

Calculated ΔC_6 constants increase regularly from He to Xe, roughly by a factor of 20, leading to the broadening constants indicated in Table II. Actually, if one looks at broadenings only, one can find a general agreement (within 35%) between experimental and theoretical constants, except for He at room temperature for which the discrepancy is practically 70%. This feature appears also when considering the temperature dependence which is observed: as $T^{0.3}$ for Ar and $T^{0.47}$ for He whereas one expects $T^{0.3}$ with this simple potential [see Eq. (1)].

The situation is much worse, however, in the case of pressure shifts since one can explain only the sense of experimental shifts. In effect, in this kind of xenon transition ($5p^55d \rightarrow 5p^56p$), $\sum\langle r^2\rangle$ is smaller in the upper level than in the lower level: Consequently, ΔC_6 constants are negative. This result can be shown not only from Aymar's calculated values of $\langle r^2\rangle$ (Ref. 20) but also from the values obtained by use of the Unsold's expression for hydrogenic atoms.[21] It results in the fact that the shifts measured here are opposite to those generally observed—toward the blue for Ne, Ar, and Xe and toward the red for He. Concerning the magnitude of these shifts, one should observe a constant ratio (2.8) between broadenings and shifts, whereas we have measured ratios between 2.3 and 7.5. Moreover, the temperature dependence is as $T^{0.6}$ for Ar and $T^{0.47}$ for He.

A partial conclusion is that, as demonstrated already,[16] broadening is more sensitive to the long-range part of the interaction, whereas shifts are more sensitive to the short-range part. With this simple potential, the agreement between experimental constants and calculated constants is better as the mass of perturbers increases: It is particularly poor in the case of He where the interaction is essentially repulsive.[22]

Introduction of a repulsive part (C_{12}/R^{12}) into the potential can lead to a more realistic form

TABLE I. T is expressed in K, γ/n and d/n are expressed in 10^{-16} MHz cm^3.[a]

Perturber	T Expt.	γ/n Expt.	d/n Expt.
He	309 ± 2	5.97 ± 0.26	−1.23 ± 0.04
Ne	301 ± 3	2.59 ± 0.18	0.34 ± 0.04
Ar	309 ± 3	4.91 ± 0.27	1.93 ± 0.08
Xe	301 ± 5	3.41 ± 0.46	1.47 ± 0.08
He	80[a] ± 4	3.18 ± 0.19	−0.65 ± 0.05
Ar	80[a] ± 4	3.25 ± 0.23	0.86 ± 0.05

[a] At zero pressure only.

TABLE II. γ/n is expressed in 10^{-16} MHz cm^3, ΔC_6 is expressed in 10^{-60} erg cm^6, and ΔC_{12} in 10^{-102} erg cm^{12}.

Perturber	γ/n Expt.	γ/d Expt.	γ/n (VDW)	ΔC_6 (VDW)	ΔC_6 (LJ)	ΔC_{12} (LJ)
He	5.97	−4.9	3.57	−29		
Ne	2.59	7.5	2.93	−56	−73	1.02
Ar	4.91	2.5	4.41	−231		
Xe	3.41	2.3	4.99	−586	−663	−1357
He	3.18	−4.9	2.38	−29		
Ar	3.25	3.8	2.94	−231		

of interaction although this last term has no real physical interpretation. According to Hindmarsch,[23] it is possible to calculate ΔC_6 and ΔC_{12} values from the determination of broadening-to-shift ratios and temperature dependences. This has been made in a number of experiments, in particular, in visible Kr lines broadened by noble gases.[24] γ/n and d/n are proportional to the functions $B(\alpha)$ and $S(\alpha)$, respectively,

$$\gamma/n = 2.59\times10^5\,(\bar{v})^{3/5}|\Delta C_6|^{2/5}B(\alpha), \tag{3}$$

$$d/n = 0.65\times10^5\,(\bar{v})^{3/5}|\Delta C_6|^{2/5}S(\alpha), \tag{4}$$

with the same units as in Eq. (1), and

$$\alpha = 0.536\,\bar{v}^{6/5}\left(\frac{\Delta C_{12}}{\hbar}\right)\left(\frac{\Delta C_6}{\hbar}\right)^{-11/5}. \tag{5}$$

$B(\alpha)$, $S(\alpha)$, and $S(\alpha)/2B(\alpha) = -2d/\gamma$ have been tabulated by Hindmarsch *et al.*[23] One can remark here that α varies with temperature as $T^{0.6}$, thus providing a critical test of the quality of interpretation.

In the case of Ne and Xe perturbers, for which we have no control by temperature, ΔC_6 and ΔC_{12} constants are easily deduced from the experimental determination of $S(\alpha)/2B(\alpha)$. The corresponding values are indicated in Table II. The ΔC_6 values found this way are very close to the ones found using a pure van der Waals potential. The ΔC_{12} value for Xe is negative, thus enhancing the attractive character of the interaction potential. In the case of He perturbers, $2d/\gamma$ is equal to -0.41 at both temperatures. This value cannot be reached on Hindmarsch's graph, the extreme value being equal to -0.3 and corresponding to a purely repulsive potential. In the case of Ar perturbers, it is possible to find representative points on the graph for both temperatures, but the variation of α with temperature is the inverse of the one expected.

A second partial conclusion is that the temperature dependence plays a major role in the interpretation of the data, even in the case of a Lennard-Jones potential which involves the adjustment of two parameters. As noted above, the Xe-He interaction seems dominated by repulsive forces, the Xe-Xe interaction by attractive forces.

IV. INTRODUCTION OF MORE REALISTIC POTENTIALS AND USE OF QUANTUM THEORY OF COLLISIONS

To have a better interpretation of our data, in particular, the temperature dependence, we have tried a new type of potential. The main part comes from the multiple-multipole interaction and can be derived from the second-order perturbation theory[25,6]

$$V(r) = \frac{e^2}{2}\sum_{k_1k_2}\frac{(2k)!}{(2k_1+1)!(2k_2)!}\,\alpha_{k_2}\langle r^{2k_1}\rangle R^{-2(k+1)}, \tag{6}$$

where k_1 and k_2 are the orders of the multipole moments, $k = k_1 + k_2$, α_{k_2} is the $2k_2$-pole moment of a ground-state perturber, and $\langle r^{2k_1}\rangle$ the mean $2k_1$ radius of the excited-state atom. In our particular case, noble gas atoms in their ground state have a much smaller size than xenon atoms in their excited states, and we can make the approximation that the terms with dipole polarizability only contribute significantly to the sum of the series. This means that we consider only $k_2 = 1$, implying that the summation of the series is easily tractable for k_1 varying from 1 to infinity. Under these conditions, expression (6) can be transformed into the following:

$$V(R) = \frac{e^2}{2}\,\frac{\alpha}{R^4}\left(1 - \frac{1}{(1-\langle r^2\rangle/R^2)^2}\right). \tag{7}$$

(Strictly speaking, this expression is correct only if we make the approximation that $\langle r^2\rangle^k \simeq \langle r^{2k}\rangle$.)

This expression diverges for $R = (\langle r^2\rangle)^{1/2}$; therefore, we use a cutoff function to compensate this divergence. The cutoff function chosen is such that it leads to simple calculations, and its validity range is narrow around the divergence. $V(R)$ is now expressed as

$$V(R) = \frac{e^2}{2}\,\frac{\alpha}{R^4}\left(1 - \frac{1}{(1-\langle r^2\rangle/R^2)^2}\right)\left\{1 - \exp\left[-\left(1 - \frac{R^2}{\langle r^2\rangle}\right)^2\right]\right\}. \tag{8}$$

In order to transform the model into an anisotropic one, as it should be, we use Unsold's hydrogenic expression for the mean-square radius,[21]

$$\langle r^2\rangle_m = \frac{\nu^2}{2}\left[5\nu^2 + 1 - 3l(l+1)\right]\frac{5l(l+1) - 3(m^2+1)}{(2l+3)(2l-1)}, \tag{9}$$

where ν is the effective principal quantum number and l and m are the orbital and magnetic quantum numbers, respectively. [By averaging expression (9) over the different levels of the $5p^5 5d$ and $5p^5 6p$ configurations of the Xe atoms, we obtain isotropic mean values of $\langle r^2\rangle$ which are in good agreement with Aymar's calculations.] The xenon noble gas quasimolecule is described in a very simple scheme where Σ, Π, and Δ components correspond to the projection of the angular momentum on the intermolecular axis (Σ corresponds to 0, Π to 1, and Δ to 2).

In the case where this projection is equal to zero, another term is added to the potential; this Fermi-type term, which is essentially repulsive, corresponds to forces between the electronic clouds of the two atoms.[26,27] It can be written as[28]

$$V_F(R) = 2\pi L'\Phi^2(R)(1-\epsilon/R), \tag{10}$$

where L' is a quantity derived from the scattering length of an electron in the vicinity of the per-

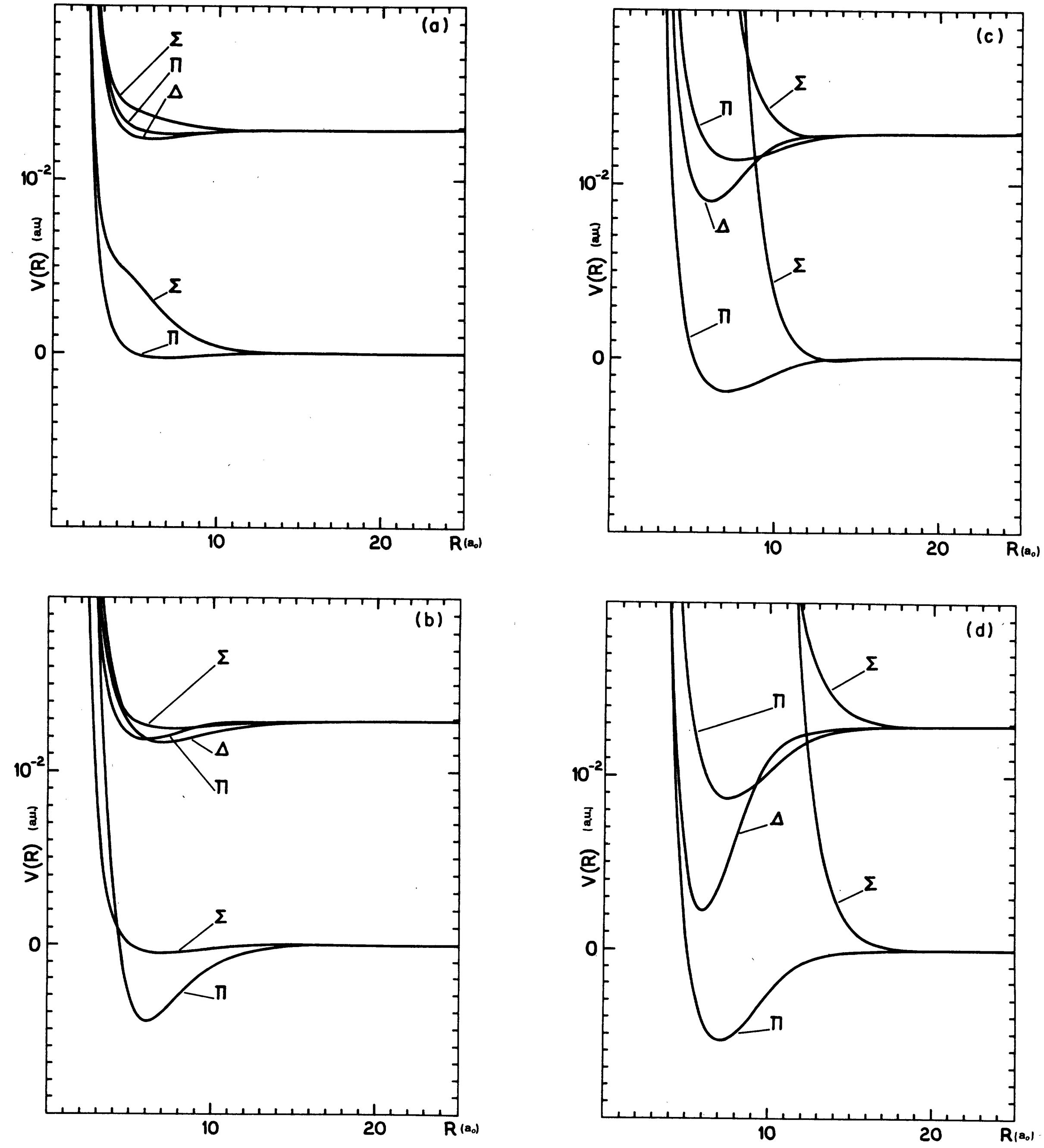

FIG. 5. (a)–(d) Potential curves for the interaction of Xe with He, Ne, Ar, and Xe, respectively. Upper curves correspond to the $5p^55d[7/2]_3$ level of xenon and lower curves to the $5p^56p[5/2]_2$. Vertical scales are in atomic units and horizontal scales in units of a_0.

turber and $\Phi(R)$ is the probability density of the optical electron in the vicinity of this perturber. ϵ/R is introduced when the scattering length becomes negative so that the Fermi potential becomes a repulsive one[28]; ϵ depends on the polarizability α of the perturber and also on the quantum number ν of the excited-state atom.

By this technique, we have calculated the interaction potentials for all couples Xe and other noble gas except Kr; the different components Σ, Π and Δ are represented in Figs. 5(a)–5(d) for the two levels of the Xe transition at $\lambda = 3.51$ μm. One can see that, as expected, the depth of the well (for Π and Δ components) is very small for He and increases regularly towards Xe whereas its position is practically constant. Σ components are only repulsive.

In our experiment, the shape of the line can be fairly well described in the frame of the impact theory by a Lorentz profile whose width and shift can be calculated from Baranger's expression.[29]

Each partial pseudomolecular transition has a width and a shift given by

$$\frac{\gamma}{n} = \frac{4\pi}{\mu}\left\langle \frac{1}{k}\sum_{l=0}^{\infty} (2l+1)\sin^2(\delta_l^i - \delta_l^f)\right\rangle , \tag{11a}$$

$$\frac{d}{n} = -\frac{\pi}{2\mu}\left\langle \frac{1}{k}\sum_{l=0}^{\infty} (2l+1)\sin 2(\delta_l^i - \delta_l^f)\right\rangle , \tag{11b}$$

where μ is the reduced mass, and δ_l^i and δ_l^f are the phase shifts, due to collisions associated with initial and final states of the pseudomolecular transition. The brackets imply an average over the relative velocity of the two atoms. In general, phase shifts are calculated by use of the Gauss-Melher quadrature[30] in the frame of the JWKB approximation[31] but here, this procedure has been employed systematically for large values of l only. For small values, phase shifts have been calculated quantitatively by resolving the Schrödinger equation (use of the Numerov method[32]). We have verified that for values of l larger than 10, the two methods lead to similar results. Total broadening and shift of the xenon line are then calculated by use of the Anderson expression.[33] In the case of broadening:

$$\gamma = \tfrac{1}{15}(2\gamma_{\Sigma\Sigma} + 3\gamma_{\Sigma\Pi} + \gamma_{\Pi\Sigma} + 3\gamma_{\Pi\Pi} + 6\gamma_{\Delta\Pi}) . \tag{12}$$

For broadenings, calculated values are systematically too large by roughly a factor of 2, independent of the temperature. This suggests the possibility of adjusting the position of the potential well [characterized by $R_0 = (\langle r^2\rangle)^{1/2}$, see expression (8)], towards $R = 0$, without modification of the long-range part of the interaction. Calculations have been performed by introducing a parameter β into expression (8), both to multiply $\langle r^2\rangle$ values and to divide the first factor. We note first a strong dependence of calculated broadenings with β values. For example, for Ar broadening at 80 K, the constant varies from 2.77×10^{-16} to 6.26×10^{-16} MHz cm^3 when β varies from 0.5 to 1. Secondly, it is possible to find a value of β, ~0.5, which gives a rather good agreement for all perturbers, except He, at the two temperatures: Corresponding values are indicated in Table III. The influence of this displacement of the potential well (here ~0.7) towards $R = 0$ has been noticed recently in other calculations using other kinds of potentials.[35,36] We can notice also that the effect is more pronounced for heavy perturbers for which the well is deeper. This suggests the possibility of adjusting also the depth of the well by adding a new parameter γ in the exponent of expression (8) but here the influence is much more reduced: For γ varying by a factor of 2, we have obtained broadening constants varying by *10% typically.* The conclusion is that, as expected, the regions of the interaction potential which correspond to long-range and medium-range collisions (those for which $R > R_0$) play a major role in the calculation of broadening constants.

TABLE III. All values are expressed in 10^{-16} MHz cm^3.

	γ/n	γ/n		d/n	d/n	
Perturber	Expt.	$\beta=1$	$\beta=0.5$	Expt.	$\beta=1$	$\beta=0.5$
He	5.97	8.72	9.11	−1.23	0.91	0.26
Ne	2.59	8.03	6.56	0.34	0.86	1.47
Ar	4.91	9.72	4.58	1.93	0.48	0.51
Xe	3.41	9.54	4.57	1.47	0.68	0.42
He	3.18	5.36	3.83	−0.65	1.44	0.93
Ar	3.25	6.26	2.77	0.86	0.51	0.92

As for shifts, the general agreement between theory and experiment is still very poor, in particular, concerning the sign of He shifts which is definitively not interpreted. One can notice also the increase of calculated Ar shifts when lowering the temperature. This disagreement is not unexpected since shifts correspond to a sum of sines [see expression 11(b)] whose variations are not monotomic, in opposition to broadenings which correspond to a sum of squared sines. This point can be shown quite generally on the real and imaginary parts of the diffusion amplitude whose variations are not correlated.

V. CONCLUSION

Measurements of the broadening constants associated with the laser line of Xe at $\lambda = 3.51$

μm were performed first in the aim of understanding a series of phenomena occuring in saturated-absorption experiments, in particular, to make evident the influence of velocity-changing collisions upon saturated-absorption profiles.[37,17] By use of the technique of linear absorption with tunable lasers, it has been possible to measure broadenings and shifts in a range of pressure where typically the impact approximation holds without restriction. Extrapolation to zero density of perturbers leads to the correct value of the natural width. A theoretical interpretation of the measurements has been attempted by use of the quantum theory of collisions.

For this interpretation we have used a potential model based on physical aspects of the collision, in a case where more than one optical electron is involved. An extensive study has been performed by testing the influence of position and depth of the potential well: It shows that the interpretation of broadenings can be rather satisfactory as soon as one displaces the position of the well toward the mass center. This point, if confirmed by other data, should be clarified, showing that potential models have to be further improved.

Calculations including both elastic and inelastic collisions should be performed also. Once these calculations are performed, one should have at his disposal a critical test for collision theory and for the exact shape of interaction potentials, in particular regarding pressure shifts whose interpretation is still unsatisfactory.

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