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DEPOLARIZATION, BROADENING AND SHIFT OF THE $\text{Rb } 5^2\text{P}_{1/2} \rightarrow 5^2\text{S}_{1/2}$ RESONANCE LINE PERTURBED BY RARE GASES

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Résumé. — On calcule les constantes de dépolarisation, d'élargissement et de déplacement de la raie de résonance du rubidium $5^2\text{P}_{1/2} \rightarrow 5^2\text{S}_{1/2}$ perturbée par tous les gaz rares dans l'approximation semi-classique en négligeant le transfert d'excitation vers le niveau $5^2\text{P}_{3/2}$ dans la gamme de température 100-1 000 K.

Les effets de trajectoire sont discutés.

Les calculs ont été effectués en utilisant les potentiels interatomiques récemment publiés par Baylis [1] ⁽¹⁾ et Pascale et Vandeplanque [2]. La comparaison des résultats théoriques et expérimentaux, quand ils existent, permet un test des potentiels et du traitement de la collision.

Abstract. — The depolarization, broadening and shift constants of the $5^2\text{P}_{1/2} \rightarrow 5^2\text{S}_{1/2}$ resonance line perturbed by rare gases are calculated in the semi-classical approximation when neglecting the excitation transfer to the $5^2\text{P}_{3/2}$ state in the energy range 100-1 000 K.

Trajectory effects are discussed.

The calculations have been performed with the interatomic potential recently published by Baylis [1] ⁽¹⁾ and Pascale and Vandeplanque [2] for all rare gases. Comparison with experiments, when available, permits a test of the potentials and the collision treatment.

1. Introduction. — Recent experimental results (Doebler and Kamke [3]) concerning the first resonance line of rubidium together with the data available for the corresponding interatomic potential curves (Baylis [1], ⁽¹⁾, Pascale and Vandeplanque [2]) have encouraged us to perform systematic calculations of the depolarization, broadening and shift constants of the $\text{Rb } 5^2\text{P}_{1/2} \rightarrow 5^2\text{S}_{1/2}$ line perturbed by all rare gases and to study their variation with energy in order to discuss the different mechanisms involved and the interatomic potentials.

2. Collision treatment. — 2.1 CHOICE OF THE APPROXIMATIONS. — The calculations were performed in the semi-classical approximation where the relative motion is classical and the internal state of the atoms is quantized.

This approximation is justified if the De Broglie wavelength $\lambda = \hbar/\sqrt{2mE}$ of the relative motion is small compared to the characteristic lengths d involved

in the problem (m is the reduced mass and E the kinetic energy)

$$\lambda \ll d. \quad (1)$$

In the case of the perturbation of a rubidium atom in its first resonant state, the mean range of the relaxation phenomena d is of the order of some atomic units (as we shall see later), the De Broglie wavelength at 400 K is of the order of 0.2 so that condition (1) is satisfied.

We must point out however that the De Broglie wavelength is obtained when neglecting the interaction potential V .

If we take it into account, $\lambda = \hbar/\sqrt{2m(E-V)}$ and then the condition (1) fails at the turning point for which the incident energy is equal to the potential energy. Moreover the classically forbidden region for which $E < V$ is not involved in our calculations. In order to determine a classical trajectory, the system should remain, during the collision, in molecular states corresponding to one potential curve.

We must now select the states involved in the problem.

We apply first the adiabatic criterion :

$$\Delta E \gg 1/\tau_c \quad (2)$$

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⁽¹⁾ Baylis, W. E., Private communication.

where we compare the energy involved in the eventual transition and the mean duration of a collision $\tau_c \sim d/v$ which is of the order of 10^{-4} a.u. for $d \sim 7$ a.u., $T \sim 400$ K and we neglect the transitions for which condition (2) is satisfied.

In the case of the ground state of Rb perturbed by a rare gas, we are thus led to expand the wave function on this state only. When we consider the first resonant state of rubidium, we can separate the fine structure states so that, in our case, we retain only the $5^2\Pi_{1/2}$ adiabatic curve.

On the other hand, for the energy gap satisfying the opposite condition

$$\Delta E \ll 1/\tau_c \quad (3)$$

we apply the sudden approximation, which is equivalent to neglecting the corresponding terms of the hamiltonian. Among these, we have in particular left

a part the splitting between the magnetic sublevels inside a given fine structure J level due to the external magnetic field and also the hyperfine coupling.

2.2 S MATRIX CALCULATION. — The S matrix calculations with the above approximations were first developed by Gordeev *et al.* [4], then fully described and somewhat improved in Roueff and Suzor [5] which will be referred to as JP1 in the following.

We adopt treatment B of JP1 in which we expand the atomic wave function $|^2P_{1/2}\rangle_{\text{Rb}} |^1S_0\rangle_{\text{RG}}$ (which is the boundary wave function of the system at $t = \pm \infty$ where RG stands for rare gas) on molecular states $|\Psi_{\pm 1/2}\rangle$ built from the adiabatic molecular states $|^2\Pi_{\pm 1/2}\rangle$ in such a way that they satisfy the symmetry of the problem (eigenfunction of the reflection operator σ_v) and that they tend to the atomic states in the limit of large R .

The corresponding S matrix is then diagonal :

$$S = \begin{bmatrix} | \Psi_{+1/2} \rangle & | \Psi_{-1/2} \rangle \\ \exp\left(-i \int_{-\infty}^{\infty} [V_{\Pi_{1/2}} + V_R] dt\right) & 0 \\ 0 & \exp\left(-i \int_{-\infty}^{\infty} [V_{\Pi_{1/2}} - V_R] dt\right) \end{bmatrix} \quad (4)$$

where $V_{\Pi_{1/2}}$ is the adiabatic potential corresponding to the $A^2\Pi_{1/2}$ adiabatic states

$$V_R = \frac{3}{4} \dot{\phi} (1 - \cos 2\chi) \quad (5)$$

where $\dot{\phi}$ is the angular velocity and χ is the acute angle defined by the relation :

$$\text{tg } 2\chi = -\frac{2\sqrt{2}}{3} \frac{(V_{\Sigma} - V_{\Pi})}{\omega + (V_{\Sigma} - V_{\Pi})/3} \quad (6)$$

V_{Σ} and V_{Π} are the molecular potentials related to the Rb (5P) interaction with a rare gas corresponding to $A=0, 1$ respectively and do not include the fine structure term. $V_{\Sigma} - V_{\Pi}$ is calculated by inverting the relations of JP1 (table II) and decreases rapidly for large internuclear distances. This treatment neglects the real transitions towards the $^2P_{3/2}$ state but takes into account some virtual transitions, namely those due to the adiabatic interaction.

This is easily seen on the following equation :

$$-i \frac{d}{dt} | \Pi_{\pm 1/2} \rangle = \left\{ \dot{\phi} J_y - i \dot{R} \frac{d}{dR} \right\} | \Pi_{\pm 1/2} \rangle \quad (7)$$

where $\dot{\phi}$ is the angular velocity and J_y is the projection of the J angular momentum on the OY axis relative to the rotating frame, which is taken to be perpendicular to the collision phase (cf. JP1). With the explicit expression of J_y , we obtain :

$$-i \frac{d}{dt} | \Pi_{\pm 1/2} \rangle = \frac{3}{4} \dot{\phi} \left(\frac{1}{3} - \cos 2\chi \right) | \Pi_{\mp 1/2} \rangle - i \sin \chi \dot{\phi} | \Pi_{\pm 3/2} \rangle + \\ + \frac{3}{4} i \dot{\phi} \sin 2\chi | \Sigma_{\mp 1/2} \rangle - i \dot{R} \frac{d\chi}{dR} | \Sigma_{\pm 1/2} \rangle. \quad (8)$$

We have only retained the first term of this expression, neglecting the short range coupling between $| \Pi_{\pm 1/2} \rangle$

and the other adiabatic states $|\Pi_{\pm 3/2}\rangle$ and $|\Sigma_{\pm 1/2}\rangle$ which tend to the $^2P_{3/2}$ atomic state of rubidium and ground state of the rare gas.

This is equivalent to a first-order perturbation treatment inside the subspace of states $|\Pi_{\pm 1/2}\rangle$, the perturbation being the non-adiabatic term.

A way of taking the virtual transitions to $|\Pi_{\pm 3/2}\rangle$, $|\Sigma_{\pm 1/2}\rangle$ into account would be to go to the second order of the perturbation. The distance between $|\Pi_{\pm 1/2}\rangle$ and the two adiabatic states $|\Pi_{\pm 3/2}\rangle$, $|\Sigma_{\pm 1/2}\rangle$ being of the order of ω (cf. JP1), we would thus induce terms of the order $\dot{\phi}^2/\omega$, R^2/ω which are small in comparison to the other terms involved, except near the turning point.

2.3 BROADENING AND RELAXATION CONSTANTS. — The disorientation cross section $\sigma_{1/2}^1$, the half width $2w$ and the shift s obtained from the impact approximation are then expressed by the following formulae [cf. JP1]

$$\sigma_{1/2}^1 = 2\sigma\left(\frac{1}{2}\frac{1}{2} \rightarrow \frac{1}{2} - \frac{1}{2}\right) = 2\pi \int_0^\infty b \frac{4}{3} \sin^2 \left[\int_{-\infty}^\infty V_R dt \right] db \quad (9)$$

where b is the impact parameter.

$\frac{1}{2} \pm \frac{1}{2}$ refers to the $|jm\rangle$ state of the Rb atom.

V_R is defined in equation (5)

$$w = 2\pi N \bar{v} \int_0^\infty b \left\{ 1 - \cos \int_{-\infty}^\infty V_R dt \cos \left[\int_{-\infty}^\infty V_{\Pi_{1/2}} dt - \int_{-\infty}^\infty V_{X^2\Sigma_{1/2}} dt \right] \right\} db \quad (10)$$

$$s = 2\pi N \bar{v} \int_0^\infty b \left\{ \cos \int_{-\infty}^\infty V_R dt \sin \left[\int_{-\infty}^\infty V_{\Pi_{1/2}} dt - \int_{-\infty}^\infty V_{X^2\Sigma_{1/2}} dt \right] \right\} db \quad (11)$$

\bar{v} is the relative mean velocity, N the density of the perturbers.

$V_{X^2\Sigma_{1/2}}$ is the molecular adiabatic potential arising from the ground $5^2S_{1/2}$ atomic state of the Rb colliding with the ground 1S_0 state of inert gases. Trajectory effects are explicitly taken into account when calculating the integrals of the form $\int_{-\infty}^\infty V dt$.

The disorientation cross section depends directly on the non-adiabatic term V_R which is the rotational coupling proportional to the angular velocity.

In the integrals involving this term V_R , the trajectory is determined by the excited adiabatic potential $V_{\Pi_{1/2}}$ and the only dependence on the velocity is indirect and arises from the effect of velocity on the trajectory $R(\varphi)$ ⁽²⁾

$$\int_{-\infty}^\infty V_R dt = \int_0^{\varphi(\infty)} d\varphi \frac{3}{4} \{ 1 - \cos 2\chi[R(\varphi)] \} . \quad (12)$$

3. The interatomic potentials. — The calculations were performed with the Baylis ⁽¹⁾ adiabatic potentials hereafter referred to as B potentials and with the Pascale and Vandeplanque [2] adiabatic potentials hereafter referred as PVP potentials.

B potentials were obtained when considering the coupling between the ground state and the first resonant state only. This coupling however should be small for most internuclear separations of interest. On the other hand, PVP potentials were determined by including many excited states in the expansion of the wave function. In our approximation (JP1) we consider that the molecular states tending to $5P$ are isolated from the others (adiabatic criterion (1)) and the formulae we give implicitly assume this fact. This

approximation can be tested by comparing the values of $V_{A^2\Pi_{3/2}}$ of PVP, which is one of the adiabatic states tending to the $^2P_{3/2}$ state of rubidium, with $V_{\Pi} + \omega$ where ω is the fine structure energy assumed to be constant. The V_{Π} molecular potential is obtained from the adiabatic potentials $V_{A^2\Pi_{1/2}}$ and $V_{B^2\Sigma_{1/2}}$ of PVP and the corresponding expression was given in table II of JP1. We found that both values agreed to within 10^{-4} for internuclear distances of interest.

Finally, we want to point out a difficulty we encountered for the long range potentials with the data given by Baylis ⁽¹⁾. The numerical values of the potentials are given up to $R = 25$ a.u. and the accuracy becomes very poor at 18 a.u. This has no influence on the relaxation cross sections, and little (10%) on the linewidth in the case of a repulsive potential. However the long range part of the potential is critical for the shifts even in the case of light rare gases when the B potential

⁽²⁾ This remark is due to one referee.

is used. Therefore, we give the results of the depolarization constant for all rare gases and only the broadening constants for He and Ne as perturbers with B potential.

4. Results and discussion. — The calculations were performed for energies ranging from 100 K up to 1 000 K and are displayed in figures 1 to 10.

In order to discuss our results, it is convenient to consider first He and Ne and then the heavy rare gases.

4.1 CASE OF REPULSIVE $V_{2\Pi_{1/2}}$ POTENTIAL (He AND Ne). — In the case of a repulsive potential, there is only one turning point which is always larger than the impact parameter and which we determine numerically.

4.1.1 Depolarization relaxation constant. — The depolarization or disorientation relaxation cross section of the $P_{1/2}$ level depends directly on the non-adiabatic potential V_R and involves an integration over the angular deflection of the internuclear axis during the collision as is shown in equation (12). In a curvilinear treatment, the total angular deflection

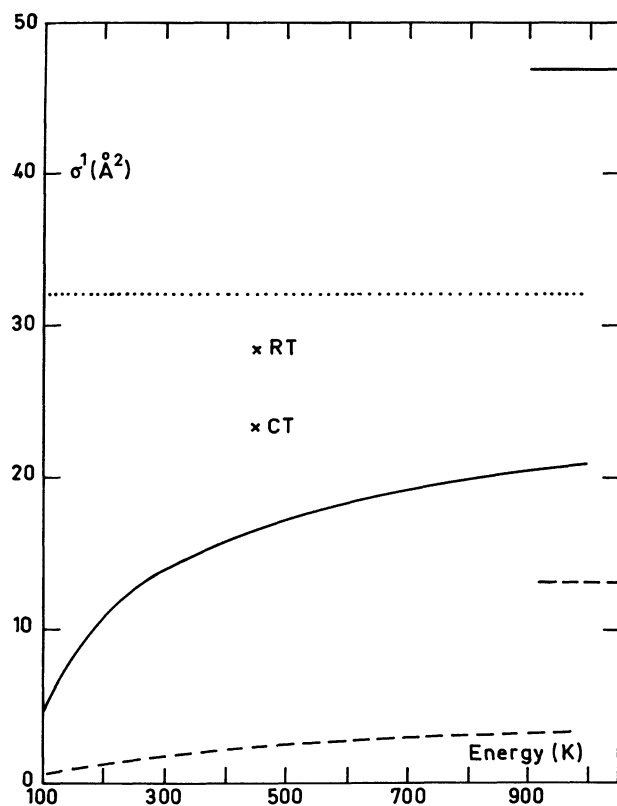


FIG. 1. — Depolarization cross-sections σ^1 (\AA^2) versus the relative kinetic energy for Rb-He. The unit of energy is K defined from the relative mean velocity : $E = \frac{4k}{\pi} T$. The results are given for a curvilinear trajectory treatment. The values corresponding to the straight line approximation are indicated at the right edge of the figure. — B potential ; ---- PVP potential ; experimental results [3]. Experimental results are given in function of temperatures reported on the same scale than the energies. Crosses refer to the previous calculations of Roueff and Suzor [5]. CT : curvilinear trajectory ; RT : rectilinear trajectory.

of the internuclear axis increases with the energy, shorter range interactions arise and the cross section increases as can be seen in figures 1 and 2.

In a rectilinear path treatment, the total angular deflection of the internuclear axis is constant and equal to π , the trajectory remains the same and the cross section does not depend on the energy. The trajectory effects in the depolarization cross section calculations are very large leading to differences of about one order of magnitude compared to the rectilinear case for helium as perturber when using the PVP (Fig. 1) potential. This effect is less pronounced with the B potential (Fig. 1).

The most recent experimental results of Doeblér and Kamke [3] ⁽³⁾ are higher than the theoretical calculations using the PVP potential by one order of magnitude and show almost no variation with energy, which may be an indication that the $A^2\Pi_{1/2}$ PVP curve is too repulsive or that $V_x - V_\pi$ is too small. The previous calculations using the exchange asymptotic potential [5] gave reasonable results compared to the experiments. It should be noted however that the experimental results do not agree with each other (see Doeblér and Kamke [3]), lying between 20 and 33 \AA^2 .

Our results in the curvilinear treatment with Neon as perturber are close when using PVP or B potentials and agree with the quantal close coupling results of Wilson and Shimoni [6] (Fig. 2) who performed their calculations with the B potential. They are also close to the experimental results of Gallagher [7], corrected by Bulos and Happer [8] and also to the experimental result of Kamke at 317 K.

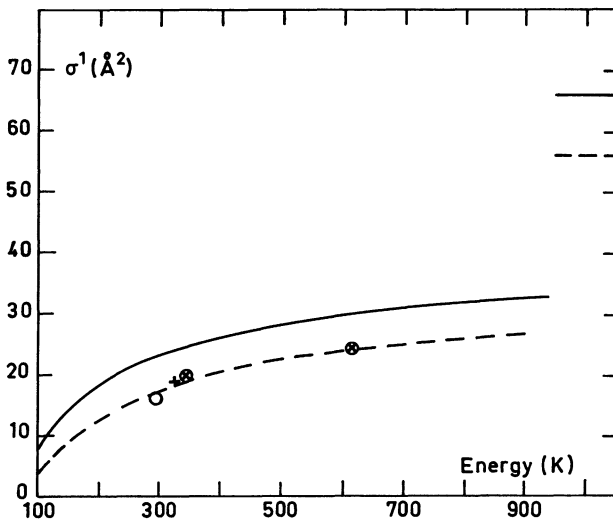


FIG. 2. — Depolarization cross-sections σ^1 (\AA^2) versus the relative kinetic energy for Rb-Ne. The notations are the same than in figure 1. \otimes : Close-coupling results of Wilson and Shimoni [6] ; \circ : Experiment of Gallagher [7] corrected by Bulos and Happer [8] ; + : Experiment of Kamke [9].

⁽³⁾ Doeblér and Kamke [3] also performed the calculations of the depolarization constants of Rb $2P_{1/2}$ perturbed by He, Kr and Xe on the same basis as we do, i.e. with PVP potential and formula (9). Comments are given in paragraph 4.2.1.

4.1.2 *Broadening and shift results.* — The variation of the halfwidth of the resonance $D_1\ 5^2P_{1/2}-5^2S_{1/2}$ line with temperature can provide a test of the interatomic potential. Whereas the broadening constants have a maximum and a minimum in the Rb-He case when using the PVP potential and a curvilinear trajectory, they increase regularly (cf. Fig. 3) with the B potential for the Rb-He couple. The results obtained in the Rb-Ne case with B and PVP potentials are close which is due to the small difference between both potentials in that case.

For a rectilinear path, we obtain a monotonic increase of the width with temperature (Fig. 3 and 4), as is usually obtained. The shifts are towards the blue for the two rare gases (Fig. 5 and 6) and exhibit small variations with the trajectory. The comparison with

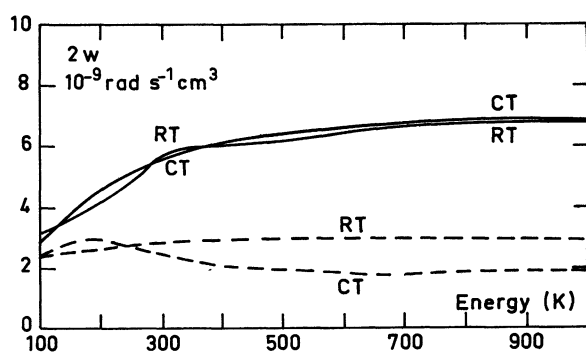


FIG. 3. — Half width of the D_1 line of Rb perturbed by He versus the relative kinetic energy. The notations are the same than in figure 1.

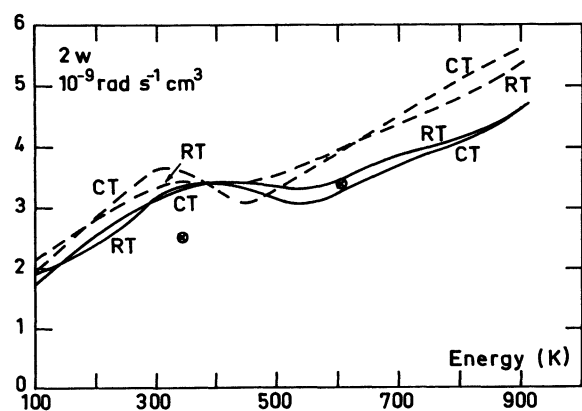


FIG. 4. — Half width of the D_1 line of Rb perturbed by Ne versus the relative kinetic energy. The notations are the same as in figures 1 and 2.

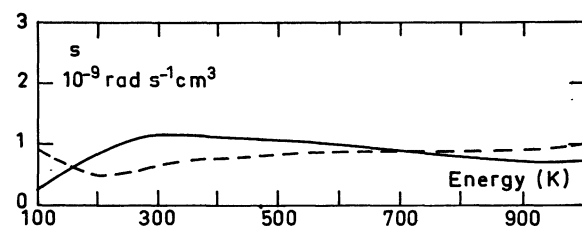


FIG. 5. — Shift of the D_1 line of Rb perturbed by He versus the relative kinetic energy using PVP potentials. — CT (curvilinear trajectory); - - - RT (rectilinear trajectory).

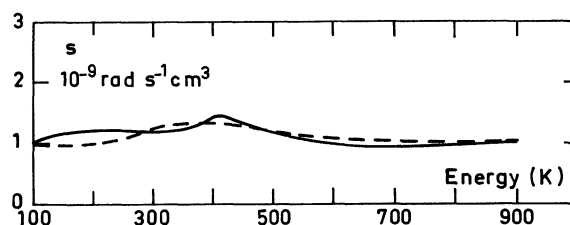


FIG. 6. — Shift of the D_1 line of Rb perturbed by Ne versus the relative kinetic energy using PVP potentials. The notations are the same as in figure 5.

the quantal close coupling results of Shimoni and Wilson [6] is shown to be satisfactory for the broadening constants.

4.2 CASE OF ATTRACTIVE $V_{II_{1/2}}$ POTENTIALS. — When attractive potentials are present, orbiting can occur, as pointed out by Doebler and Kamke [3], and is the dominant effect.

4.2.1 *Disorientation relaxation constants.* — The disorientation relaxation cross-sections for the three heavy rare gases as perturbers were calculated with the PVP and B potentials, and are displayed in figure 7. When using the PVP potential, the cross sections increase from Ar to Xe which is due to the enhancement of the potential depth (thus of the orbiting phenomena) and of the long range attractions.

The occurrence of a maximum in the temperature variation is due to the competition between the attraction and the centrifugal repulsion, which becomes predominant at high temperature, as already pointed out in Doebler and Kamke [3]. However, we have to point out an error in the calculations of Doebler and Kamke [3, 10] for xenon as perturber. It turns out that in this case, the discrepancy between theoretical and experimental results is removed (Fig. 8)

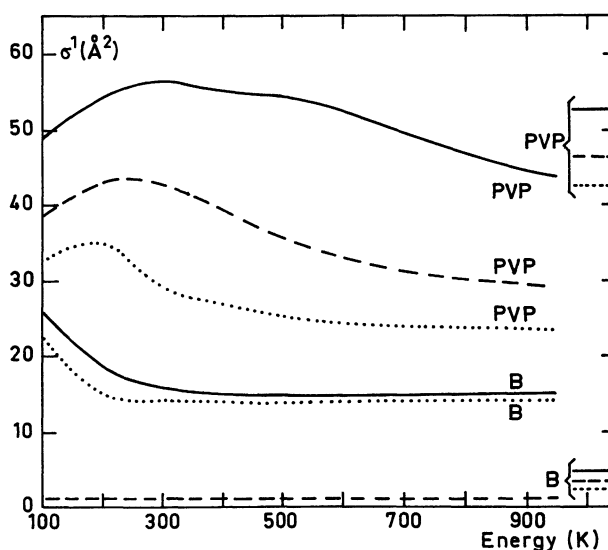


FIG. 7. — Depolarization cross-sections σ^1 (\AA^2) for the heavy rare gases versus the relative kinetic energy. Ar; - - - Kr; — Xe. The used potentials are on the curves. Straight line results are given at the right edge of the figure.

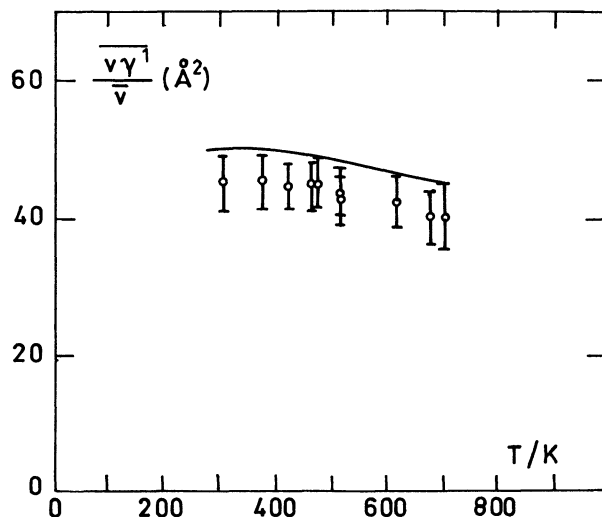


FIG. 8. — Comparison between theoretical and experimental results of the depolarization cross-section for Rb-Xe using PVP potentials versus temperature. I experiment of Doeblner and Kamke [3]; — our results.

and that the PVP potential seems quite reliable for Rb-Kr and Rb-Xe. The results are quite different and considerably reduced when B potentials are involved as can be seen in figure 7. This is principally due to the different well depths of the $A^2\Pi_{1/2}$ potentials for both potentials shown in table I.

TABLE I

Potential well depths of $V_{\Pi_{1/2}}$ for Rb-heavy rare gas couples in KK for PVP and B potentials (1 KK = 1 000 cm^{-1})

Ar		Kr		Xe	
PVP	B	PVP	B	PVP	B
0.164	0.066	0.335	0.079	0.544	0.125

Orbiting still occurs for Ar and Xe but, surprisingly, not in the case of Kr. This can be explained by a very smooth variation of the potential after the well depth so that the maximum of the centrifugal barrier occurs at too great an internuclear distance when V_R is negligible.

From the comparison with the experiments of Doeblner and Kamke [3], the B potentials seem to be too shallow for Kr and Xe.

Data for Ar are unfortunately missing.

4.2.2 Broadening and shift results. — In the broadening and shift calculations, the phase shifts

$$\int_{-\infty}^{\infty} V_{\Pi_{1/2}} dt \quad \text{and} \quad \int_{-\infty}^{\infty} V_{X^2\Sigma_{1/2}} dt$$

are subject to huge variations over a relatively large range of impact parameters when orbiting occurs so that the corresponding integrands oscillate very rapidly and the integrals are almost averages over the

intervals involved and depend very little on the exact form of the potential.

An interesting feature of figure 9 is the inversion of the inequality between the different half widths.

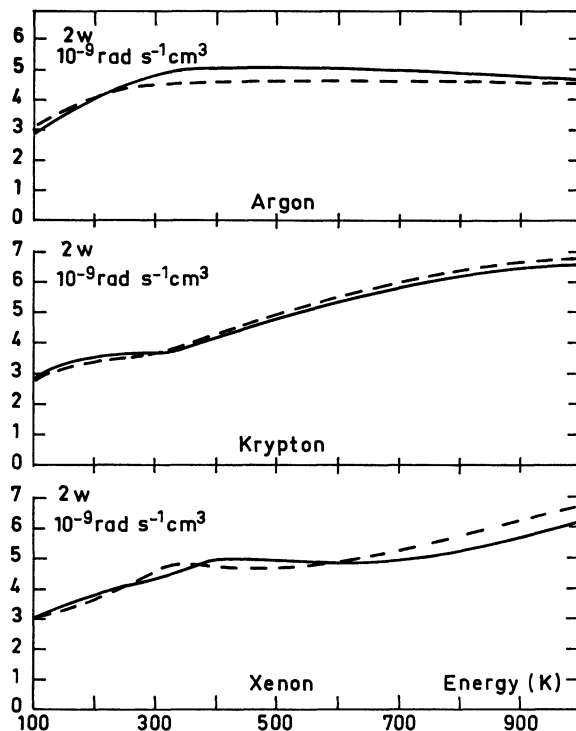


FIG. 9. — Half width of the D_1 line of Rb perturbed by the heavy rare gases versus the relative kinetic energy using PVP potentials. The notations are the same as in figure 5.

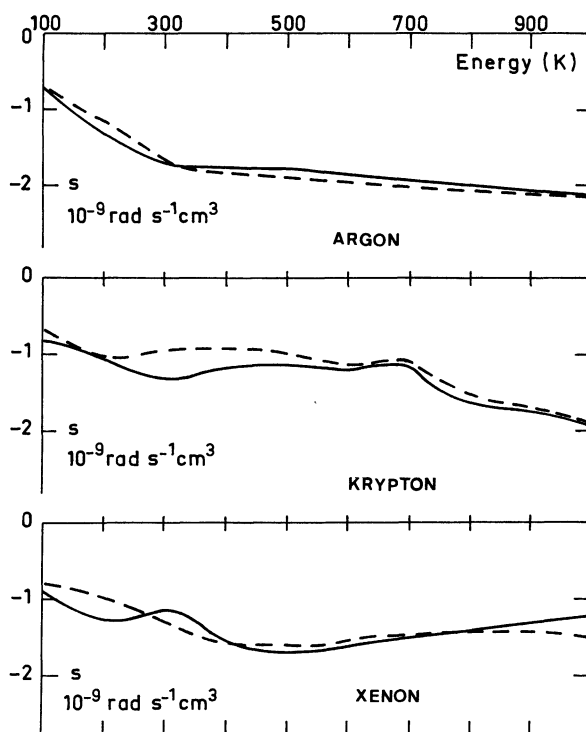


FIG. 10. — Shifts of the D_1 line of Rb perturbed by the heavy rare gases versus the relative kinetic energy using PVP potentials. The notations are the same as in figure 5.

At low temperature, we have $w_{Kr} < w_{Xe} < w_{Ar}$; at high temperature, the relation is inverted. The shifts are displayed in figure 10 and are towards the red.

5. Conclusion. — Apart from the calculations concerning Rb-He performed with the PVP potential, our results agree reasonably well with the existing experiments. Moreover, they reproduce with considerable gain of computing time the available quantal calculations. This supports the usefulness of the semi-classical approximation in the energy range considered and also the reliability of the PVP potential except in the Rb-He case. The results obtained with B potentials are more satisfactory than with PVP in the case of Rb-He but still do not agree with experiments.

The results obtained for Rb-Ne with both potentials are very close and reasonable. The B potentials for the heavy rare gases seem too shallow.

Trajectory effects are discussed and we show that they are especially critical for the depolarization of the D_1 line induced by collisions with light rare gases. Experiments relative to broadening and shift measurements are unfortunately missing although their variation with temperature can exhibit interesting features.

From $T = 400$ K a new channel is open towards the $^2P_{3/2}$ level. However, when the total energy is

of the order of the threshold, the semi-classical approximation cannot take account of the change of the relative velocity during the collision and it is of no help to solve the six coupled differential equations relative to the P - alkali - rare gas couples (see for example, Masnou and Roueff [11]) when both fine structure states are treated together. Only the quantal treatment is then valid. However, the agreement between our results for $E = 615$ K with those of Wilson and Shimoni [6] seems to justify the validity of our assumptions.

Acknowledgments. — The authors would like to thank Doctor B. Kamke for fruitful discussions and communication of her results before publication.

They are also indebted to the referees for interesting suggestions and remarks.

Note added in proof. — New quantal calculations on the depolarization cross section of the D_1 line of Rubidium perturbed by He and Xe [12] just appeared. The authors used the B potential. The agreement between quantal and semi-classical results is obtained when he is concerned. However, the quantal results are about twice our semi-classical values when Xenon is the perturber. Further investigation is needed on this point.

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