

The Impact Parameters of the Broadening and Shift of Spectral Lines

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Abstract. The classical theory of collisional broadening and shift coefficients (β , δ) of an isolated spectral line was used to obtain simple analytical formulae for calculating both β and δ . These formulae were obtained on the assumption that the short range interaction was effective only in the broadening, while the long range was effective in the shift of the spectral line. These coefficients, β and δ depended on the limiting phase shifts responsible for broadening η_{ob} and shift η_{os} . It was found that the values of η_{ob} and η_{os} were not equal to each other, as was proposed by Weisskopf $\eta_{ob} = \eta_{os} = 1$, but were instead $\eta_{ob} = \pm \pi / 5$ and $\eta_{os} = \pm \pi / 2$. The correct signs of these phases were obtained and defined. When these phases were applied with their correct signs in the approximate formulae, the broadening β_c and shift δ_c coefficients for some interactions of Tl, Hg, Cd, Zn, Ar and Ne with inert gases and self-interactions were in agreement with the corresponding values obtained numerically by other authors. The limit at which the shift changed its sign was also obtained. New impact parameters which were not known up to now have been discussed and obtained.

Keywords: spectral line, broadening coefficient β , shift coefficient δ , impact parameters, spectral line shift, spectral line broadening

Introduction

It has long been appreciated that studying the collision broadening and shift of spectral lines contain information concerning the interatomic potentials between the radiating and perturbing atoms. The theoretical treatment of this process is of great interest for the region of low densities at which the interactions of two particles are predominant, where the impact approximation takes place, and the half-width and shift are proportional to the density of the perturbing gas. In this case, the broadening and shift coefficients β and δ , respectively, are specified for such interactions. Quantum mechanical formulation of the impact approximation to the theory of collision broadening and shift of spectral lines gives results which differ little from those of the classical theory. It seems worthwhile, therefore, to use the classical theory of collision broadening and shift developed for any interaction potential. To interpret the experimental data, the theoretical values of broadening and shift parameters in the impact limit of line broadening theory have been calculated for van der Waals and Lennard-Jones potentials by many authors (Dygdala *et al.*, 1989; Dygdala, 1988; Bielski *et al.*, 1985; Czychaj and Sienkiewicz, 1984). The results of these calculations were obtained by the numerical solution of the Lindholm and Foley impact theory of broadening developed by Hindmarsh *et al.* (1967) and Helmi (1994), who concluded that the impact parameters for the broadening and shift coefficients of spectral lines must be different, contrary to the proposed value $\rho_{\beta} =$

$\rho_{\delta} = \rho_0$ due to the Weisskopf phase shift $\eta_0 = 1$ (Helmi, 1994), which has no basis. Helmi and Roston (2000) obtained simple analytical formulae for calculating β and δ in case of Lennard-Jones potential. These formulae were based on the assumption that the phase shifts for the broadening η_{ob} and shift η_{os} were different. These values are given by $\eta_{ob} = \pm \pi / 5$ and $\eta_{os} = \pm \pi / 2$. Comparing the calculated values of β and δ with that calculated by the numerical method (Hindmarsh *et al.*, 1967), it was found that there was a good agreement between the two values for some interactions, when η_{ob} and η_{os} were positive and when other interactions were negative.

The aim of this work was to obtain the correct sign of the broadening and shift phases η_{ob} and η_{os} and how to apply these to calculate the values of the new impact parameters, ρ_{ob} and ρ_{os} , responsible for the broadening and shift of spectral line, and also to obtain the critical value of the impact parameter, ρ_s , which separates between the red and blue shifts of the spectral line.

Theoretical Background

According to the adiabatic phase shift theory, the broadening β and shift δ coefficients are given by:

$$\beta = 4 \pi \hat{v} \int_0^{\infty} \rho [1 - \cos \eta(\hat{v}, \rho)] d\rho \quad (1a)$$

$$\delta = 2 \pi \hat{v} \int_0^{\infty} \rho \sin \eta(\hat{v}, \rho) d\rho \quad (1b)$$

where:

$\eta(\hat{v}, \rho)$ = the total phase shift caused by a single collision occurring at the impact parameter ρ and relative velocity \hat{v} .

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If the perturber follows a straight line trajectory, $\eta(\hat{v}, \rho)$ can be written as (Findeisen *et al.*, 1987):

$$\eta(\hat{v}, \rho) = \frac{2}{\hbar \hat{v}} \int_0^{\infty} \frac{R \Delta V(R)}{[R^2 - \rho^2]^{1/2}} dR \quad (2)$$

where:

R = the interatomic separation

$\Delta V(R)$ = the difference between the adiabatic potentials describing the interaction between the perturber and emitting atom in its upper and lower states

For the simplest case of monatomic inverse-power potentials $\Delta V(R) = \hbar C_n R^{-n}$, the phase shift (\hat{v}, ρ) takes the form:

$$\eta(\hat{v}, \rho) = \frac{\alpha_n C_n}{\hat{v} \rho^{n-1}} \quad (3)$$

where:

$$\alpha_n = \sqrt{\pi} \frac{\Gamma(n-1)/2}{\Gamma(n/2)} \quad (4)$$

The approximate formulae for β and δ . If we assume that the effective part for broadening β in (1a) comes from the near distances ($0 - \rho_{ob}$) and the effective part for the shift δ in (1b) comes from the far distances ($\rho_{o\delta} \rightarrow \infty$), then (1a) and (1b) will take the form:

$$\beta = 4 \pi \hat{v} \int_0^{\rho_{ob}} \rho [1 - \cos \eta(\hat{v}, \rho)] d\rho \quad (5a)$$

$$\delta = 2 \pi \hat{v} \int_{\rho_{o\delta}}^{\infty} \rho \sin \eta(\hat{v}, \rho) d\rho \quad (5b)$$

It may be seen from (3) that $\eta(\hat{v}, \rho)$ will be large in the short range ($0 - \rho_{ob}$) and $\cos \eta(\hat{v}, \rho)$ is quickly oscillating, so that $\int \cos \eta(\hat{v}, \rho) = 0$, while $\eta(\hat{v}, \rho)$ will be very small in the long range ($\rho_{o\delta} \rightarrow \infty$), so that $\sin \eta(\hat{v}, \rho) \approx \eta(\hat{v}, \rho)$. From this was found that (Helmi and Roston, 2000):

$$\beta = 2 \pi \hat{v} \int_0^{\rho_{ob}} \rho d\rho = 2 \pi \rho_{ob}^2 \quad (6a)$$

$$\delta = 2 \pi \hat{v} \int_{\rho_{o\delta}}^{\infty} \rho \eta(\hat{v}, \rho) d\rho \quad (6b)$$

where:

ρ_{ob} and $\rho_{o\delta}$ = respectively, the broadening and shift impact parameters

Applications to potentials. Different types of inverse-power potentials. The approximate formulae (6) are used to express β and δ in terms of the limiting values of impact parameter ρ , and hence, via (3), the limiting values of η . Introducing the van der Waal's difference potential $\Delta V(R) = \hbar C_n R^{-n}$ in (2), the phase

parameters due to the broadening η_{ob} and shift $\eta_{o\delta}$ are given as (Helmi and Roston, 2000):

$$\eta_{ob} = [2 \int_0^{\infty} x [1 - \cos x^{1-n}] dx]^{\frac{1-n}{2}} \quad (7a)$$

$$\eta_{o\delta} = [(n-3) \int_0^{\infty} x \sin x^{1-n} dx]^{\frac{1-n}{3-n}} \quad (7b)$$

The average values of η_{ob} and $\eta_{o\delta}$ for different values of n [$n = 3, 4$ and 6] were calculated by Helmi and Roston (2000), which gives $\eta_{ob} = \pm \pi / 5$ and $\eta_{o\delta} = \pm \pi / 2$.

Lennard-Jones potential. The Lennard-Jones difference potential between the excited and the ground states of the quasimolecule consisting of radiating and perturbing atoms is given by:

$$\Delta V(R) = \hbar \Delta C_{12} R^{-12} - \hbar \Delta C_6 R^{-6} \quad (8)$$

where:

R = the distance between the colliding atoms

ΔC_6 , ΔC_{12} = constants, depending on the states of these atoms

The pressure broadening β and shift δ coefficients in the case of L-J potential are given as (Hindmarsh *et al.*, 1967):

$$\beta = 8 \pi \left(\frac{3\pi}{8} \right)^{\frac{2}{5}} \hat{v}^{\frac{3}{5}} (\Delta C_6)^{\frac{2}{5}} B(\alpha) \quad (9a)$$

$$\delta = 2 \pi \left(\frac{3\pi}{8} \right)^{\frac{2}{5}} \hat{v}^{\frac{3}{5}} (\Delta C_6)^{\frac{2}{5}} S(\alpha) \quad (9b)$$

where:

the broadening and shift functions $B(\alpha)$ and $S(\alpha)$ are defined by the following integrals:

$$B(\alpha) = \int_0^{\infty} x \sin^2 \frac{1}{2} (\alpha x^{11} - x^5) dx \quad (10a)$$

$$S(\alpha) = \int_0^{\infty} x \sin (\alpha x^{11} - x^5) dx \quad (10b)$$

where:

$$\alpha = 0.536 \hat{v}^{\frac{6}{5}} (\Delta C_6)^{-\frac{11}{5}} \Delta C_{12}$$

The functions $B(\alpha)$ and $S(\alpha)$ were obtained numerically for some chosen values of α . The phase shift $\eta(\hat{v}, \rho)$ in case of L-J potential is given by introducing (8) into (2) as:

$$\eta(\hat{v}, \rho) = \left(\frac{\alpha_{12} \Delta C_{12}}{\hat{v}} \right) \rho^{-11} - \left(\frac{\alpha_6 \Delta C_6}{\hat{v}} \right) \rho^{-5} \quad (11)$$

Introducing the obtained average values $\eta_{ob} = \pm 0.63$ and $\eta_{o\delta} = \pm 1.57$ into (11), the broadening and shift impact parameters ρ_{ob} and $\rho_{o\delta}$ in the case of L-J potential are obtained as:

$$\left(\frac{\alpha_{12} \Delta C_{12}}{\hat{v}} \right) \rho_{ob}^{-11} - \left(\frac{\alpha_6 \Delta C_6}{\hat{v}} \right) \rho_{ob}^{-5} = \pm \frac{\pi}{5} = \pm 0.63 \quad (12a)$$

$$\left(\frac{\alpha_{12} \Delta C_{12}}{\hat{v}} \right) \rho_{0\delta}^{-11} - \left(\frac{\alpha_6 \Delta C_6}{\hat{v}} \right) \rho_{0\delta}^{-5} = \pm \frac{\pi}{2} = \pm 1.57 \quad (12b)$$

The positive sign in (12a, b) denotes that the repulsion forces are more than the attraction forces, while the negative sign denotes that the attraction forces are more than the repulsive forces at the collision time.

To obtain the broadening β and shift δ coefficients in case of L-J potential, we introduce (11) into (6a, b), which gives:

$$\beta = \hat{v} \rho_{0b}^2 \quad (13a)$$

$$\delta = \hat{v} \left(\frac{\alpha_{12} \Delta C_{12}}{9 \hat{v}} \right) \rho_{0\delta}^{-9} - \left(\frac{\alpha_6 \Delta C_6}{3 \hat{v}} \right) \rho_{0\delta}^{-3} \quad (13b)$$

Knowing ρ_{0b} and $\rho_{0\delta}$ from (12a, b), β and δ can be obtained directly from the approximated formulae (13a, b).

Applying the Lennard-Jones (L-J) potential with the same obtained average values of η_{0b} and $\eta_{0\delta}$, and comparing the results obtained by the approximated formulae (6a, b) for β and δ with the results obtained for these coefficients by numerical calculations of (Hindmarsh *et al.*, 1967), Helmi and Roston (2000) found that there was an agreement between the two results for some interactions when η_{0b} and $\eta_{0\delta}$ were positive, and other interactions were negative, but Helmi and Roston (2000) did not clarify the reason of the different signs, and as to when these signs could be applied.

To obtain the necessary sign used in (12a, b), the following was proceeded:

Let ρ_0 , ρ_E , ρ_{0b} and $\rho_{0\delta}$ be the impact parameters corresponding, respectively, to the phase shifts $\eta = 0$, $\eta = \eta_E$ (the phase shift well-depth), $\eta_{0b} = \pm 0.63$ and $\eta_{0\delta} = \pm 1.57$. When $\eta(\hat{v}, \rho)$ given by (11) was plotted against ρ for different interactions, the plotted curves had the form shown in Fig. 1-3. The parameters ρ_0 , ρ_E and η_E are given from (11) as:

$$\rho_0 = \left[\frac{21 \Delta C_{12}}{32 \Delta C_6} \right]^{1/6}, \quad \rho_E = 1.14 \rho_0$$

$$\eta_E = \left(\frac{\alpha_{12} \Delta C_{12}}{V} \right) \rho_E^{-11} - \left(\frac{\alpha_6 \Delta C_6}{V} \right) \rho_E^{-5} \quad (14)$$

It may also be seen from (13b) that the impact parameter ρ_δ , which separates between the negative and positive signs of the shift coefficients δ , is given by:

$$\rho_\delta = \left[\frac{7 \Delta C_{12}}{32 \Delta C_6} \right]^{1/6} = 1.2 \rho_0 \quad (15)$$

So that if $\rho_{0\delta} < \rho_\delta$ in (12b), then δ has a positive sign and the spectral line will be shifted to the blue wing, but if $\rho_{0\delta} > \rho_\delta$,

then δ has a negative sign and the spectral line will be shifted to the red wing.

Results and Discussion

To obtain the appropriate sign of $\eta_{0b} = \pm 0.63$ and $\eta_{0\delta} = \pm 1.57$, we proceed as follows:

knowing, ΔC_6 , ΔC_{12} and \hat{v} , η_E can be obtained using (14);

(1) if $\eta_E > -0.63$

then:

ρ_{0b} and $\rho_{0\delta}$ can be obtained using the positive sign for η_{0b} and $\eta_{0\delta}$ according to (12a, b), in this case $\rho_{0\delta} < \rho_{0b} < \rho_E$ (Fig. 1),

(2) if $\eta_E \leq -1.57$

then:

ρ_{0b} and $\rho_{0\delta}$ will be taken with the negative sign for η_{0b} and $\eta_{0\delta}$, in this case $\rho_{0b} > \rho_{0\delta} > \rho_E$ (Fig. 2),

(3) if $\eta_E < -0.63$, but > -1.57

then:

ρ_{0b} will be taken with the negative sign for η_{0b} , so that, $\rho_{0b} > \rho_E$, however, $\rho_{0\delta}$ will be taken with the positive sign for $\eta_{0\delta}$, so that $\rho_{0\delta} < \rho_E$ (Fig. 3).

The calculated coefficients β_c and δ_c for different interactions are illustrated in Table 1, with the corresponding Hindmarsh values β_H and δ_H , for the interactions of Ar, Ne, Tl, Hg, Cd and Zn atoms with the inert gases Xe, Kr, Ar, Ne and He.

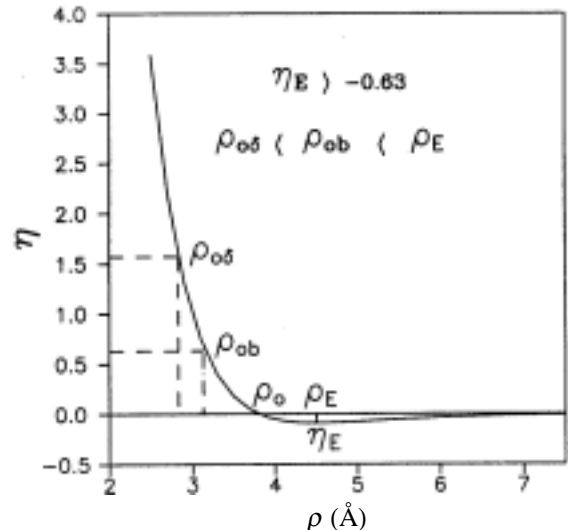


Fig. 1. The phase shift η as a function of the impact parameter ρ , when $\eta_E > -0.63$.

Table 1. The calculated values of pressure broadening β_c and shift δ_c coefficients with the corresponding Hindmarsh values β_H and δ_H in units $10^{-20}/\text{cm}/\text{atom}/\text{cm}^3$ for Ar, Ne, Tl, Hg, Cd and Zn perturbed by inert gases*

| Perturber | Parameters | | Phase | Impact parameters | | | | Broadening | | Shift potential | |
|-------------------------------------|--------------|-----------------|----------|-------------------|----------|-------------|------------------|------------|-----------|-----------------|------------|
| | ΔC_6 | ΔC_{12} | η_E | ρ_δ | ρ_E | ρ_{ob} | $\rho_{o\delta}$ | β_H | β_C | δ_H | δ_C |
| Ar line 703 nm, T = 330 K | | | | | | | | | | | |
| Ar[1] | 531.8 | 83750 | -0.060 | 18.0 | 24.7 | 18.8 | 17.6 | 7.040 | 6.930 | 0.200 | 0.200 |
| Ne[1] | 130.0 | 55870 | -0.005 | 21.3 | 29.2 | 18.4 | 17.0 | 8.250 | 8.170 | 0.990 | 1.000 |
| He[1] | 70.50 | 48190 | -0.001 | 23.0 | 31.7 | 17.3 | 16.0 | 13.89 | 13.75 | 1.890 | 1.780 |
| Ne line 540 nm, T = 330 K | | | | | | | | | | | |
| Ne[4] | 5.400 | 1.2400 | -0.100 | 6.09 | 8.34 | 6.52 | 6.15 | 1.153 | 1.170 | -0.028 | -0.028 |
| He[4] | 2.900 | 0.8800 | -0.025 | 6.37 | 8.73 | 6.22 | 5.79 | 1.870 | 1.870 | 0.150 | 0.150 |
| Tl line 377.68 nm, T = 860 K | | | | | | | | | | | |
| Xe[2,3] | 145.2 | 30.910 | -5.290 | 5.99 | 8.21 | 14.1 | 11.7 | 3.270 | 3.160 | -1.100 | -1.170 |
| Kr[2,3]] | 49.40 | 20.610 | -1.900 | 6.08 | 9.49 | 12.4 | 10.0 | 2.840 | 2.840 | -1.400 | -1.100 |
| Ar[2,3] | 58.70 | 15.250 | -1.170 | 6.20 | 8.49 | 7.72 | 7.12 | 1.900 | 1.949 | -1.180 | -1.200 |
| Ne[2,3] | 14.30 | 6.5800 | -0.130 | 6.82 | 9.34 | 7.42 | 7.00 | 1.640 | 1.800 | -0.080 | -0.090 |
| Hg line 253.6 nm, T = 860 K | | | | | | | | | | | |
| Hg[5] | 11.700 | 0.0572 | -10.98 | 3.19 | 4.38 | 8.76 | 7.29 | 1.079 | 1.085 | -0.396 | -0.394 |
| Xe[6] | 0.2970 | 0.0010 | -0.270 | 3.01 | 4.28 | 3.52 | 3.36 | 0.207 | 0.202 | -0.037 | -0.036 |
| Kr[7] | 0.7920 | 0.0080 | -0.300 | 3.61 | 4.94 | 4.08 | 3.93 | 0.304 | 0.314 | -0.068 | -0.067 |
| Ar[8] | 0.0630 | 0.4970 | -0.003 | 5.09 | 6.97 | 4.20 | 3.87 | 0.438 | 0.443 | 0.055 | 0.059 |
| Ne[8] | 0.7450 | 0.0640 | -0.027 | 5.16 | 7.05 | 5.07 | 4.72 | 0.850 | 0.847 | 0.064 | 0.066 |
| Cd line 326.1 nm, T = 860 K | | | | | | | | | | | |
| Cd[9] | 7.2200 | 0.0451 | -4.080 | 3.33 | 4.56 | 7.45 | 6.15 | 1.140 | 1.070 | -0.365 | -0.396 |
| Xe[10] | 1.3900 | 0.0110 | -0.670 | 3.46 | 4.75 | 4.50 | 3.90 | 0.359 | 0.370 | -0.154 | -0.156 |
| Kr[11] | 0.2190 | 0.0127 | -0.018 | 4.85 | 6.61 | 4.58 | 4.26 | 0.428 | 0.437 | 0.039 | 0.041 |
| Ar[10] | 0.2197 | 0.0306 | -0.458 | 3.82 | 5.21 | 4.37 | 4.22 | 0.549 | 0.510 | -0.180 | -0.175 |
| Ne[10] | 0.3740 | 0.0388 | -0.760 | 3.63 | 4.96 | 4.63 | 4.10 | 0.775 | 0.753 | -0.355 | -0.370 |
| He[10] | 0.3170 | 0.0106 | -0.010 | 4.43 | 6.03 | 4.04 | 3.75 | 1.190 | 1.200 | 0.123 | 0.127 |
| Zn line 307.6 nm, T = 860 K | | | | | | | | | | | |
| Zn[12] | 1.7875 | 0.0199 | -0.475 | 3.67 | 5.02 | 4.22 | 4.07 | 0.491 | 0.441 | -0.163 | -0.161 |
| Xe[13] | 10.532 | 0.1926 | -2.558 | 3.98 | 5.46 | 9.36 | 7.71 | 1.220 | 1.188 | -0.665 | -0.705 |
| Kr[14] | 2.6630 | 0.0028 | -5.369 | 2.47 | 3.39 | 5.91 | 4.95 | 0.829 | 0.817 | -0.268 | -0.291 |
| Ar[14] | 0.1650 | 0.0072 | -0.012 | 4.61 | 6.31 | 4.25 | 3.95 | 0.514 | 0.515 | 0.051 | 0.053 |
| Ne[15] | 0.1337 | 0.0030 | -0.014 | 4.12 | 5.63 | 3.83 | 3.56 | 0.529 | 0.531 | 0.052 | 0.054 |

* the values of ρ are in \AA units; ΔC_6 in units $10^{-32} \text{cm}^6 \text{rad s}^{-1}$, and ΔC_{12} in units $10^{-74} \text{cm}^{12} \text{rad s}^{-1}$, which are taken from the references given below:

[1] Bielski *et al.* (1985), [2] Dygdala (1988), [3] Dygdala *et al.* (1989), [4] Bielski *et al.* (1980), [5] Czuchaj *et al.* (1997), [6] Okunishi *et al.* (1990), [7] Kurosawa *et al.* (1998), [8] Petzold and Behmenburg (1978), [9] Helmi *et al.* (1996), [10] Czuchaj and Stoll (1999), [11] Czajkowski *et al.* (1991), [12] Czajkowski and Koperski (1999), [13] Wallace *et al.* (1991), [14] Wallace *et al.* (1992), [15] Koperski and Czajkowski (2000)

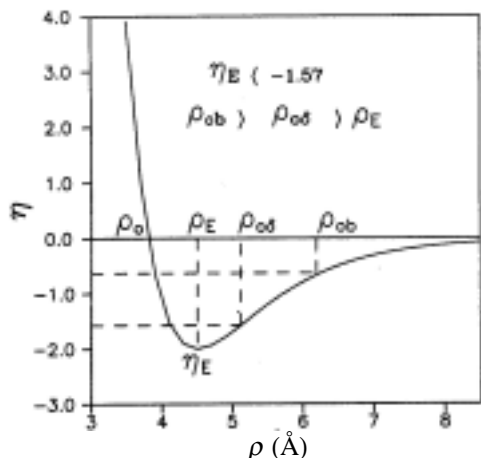


Fig. 2. The phase shift η as a function of the impact parameter ρ , when $\eta_E \leq -1.57$.

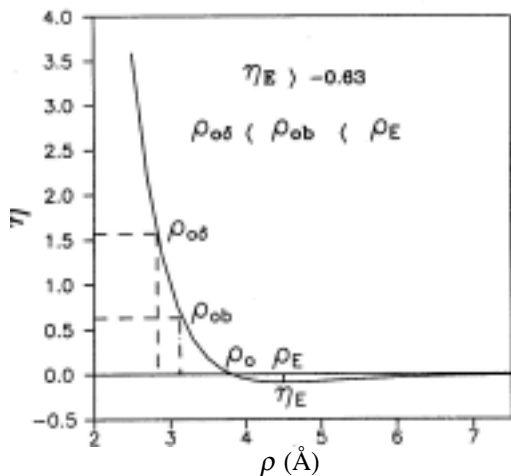


Fig. 3. The phase shift η as a function of the impact parameter ρ , when $-1.57 < \eta_E < -0.63$.

Conclusions

The following conclusions are based on the fact that the calculated coefficients β_c and δ_c by simple analytical formulae obtained by the present authors, when the Lennard-Jones potential was applied, are in good agreement as shown in Table 1, with the corresponding coefficients obtained numerically by other authors. This has led to the following considerations:

- (1) new impact parameters ρ_o , ρ_E , ρ_{ob} , $\rho_{o\delta}$ and ρ_{δ} , which are firstly defined and obtained,
- (2) the impact parameters ρ_{ob} , and $\rho_{o\delta}$ responsible, respectively, for the broadening and shift of spectral lines are different, and depend strongly on the values of ΔC_6 and ΔC_{12} , the values and signs of the phase shifts η_{ob} and $\eta_{o\delta}$ due to the broadening and shift,

- (3) the phases η_{ob} and $\eta_{o\delta}$ at which the broadening and shift, respectively, start to take place are also different in values, which are the same for all interactions, $\eta_{ob} = \pm 0.63$ and $\eta_{o\delta} = \pm 1.57$,
- (4) the sign of phases η_{ob} or $\eta_{o\delta}$ depended on the value of the phase η_E at the equilibrium position of phases, which value is given by (14),
- (5) the impact parameter ρ_{δ} which separates between the positive and negative signs of the shift of spectral line was obtained and given by a simple formula (15), so that if $\rho_{o\delta} < \rho_{\delta}$, then the line was shifted to the blue wing ($\delta =$ positive value), but if $\rho_{o\delta} > \rho_{\delta}$, then it was shifted to the red wing ($\delta =$ negative value),
- (6) as the calculated coefficients β_c and δ_c , using the approximated formulae, are in good agreement with those obtained before by numerical calculations, then all assumptions leading to the approximated formulae are valid and the obtained formulae, furthermore, can be easily used with other complicated potentials.

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