
Long-Range Interactions and the Oscillator Strength for the ${}^11_u, {}^10_u^+ \leftarrow X{}^10_g^+$ Transitions in Zn_2

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The interaction potentials for Zn_2 electronic states 11_u and ${}^10_u^+$ correlated with $(4{}^1P_1 + 4{}^1S_0)$ asymptote in the long-range region (10–25) Å and the oscillator strength f for the absorption transition to these states are derived experimentally from a careful analysis of the self-broadened resonance 213.8 nm line of Zn. It is found that although the interatomic interactions at so long distances are dominated by resonance potential of the form C_3/R^3 resulting in the Lorentzian line shape, the parameter C_3 and thus f value significantly decrease with R decreasing. This effect proved to be a linear function of R^{-1} yielding the oscillator strength for the considered line $f_0 = 1.456 \pm 0.010$ from $R \rightarrow \infty$ extrapolation. This value seems to be the most accurate experimental result as yet and shows that the best theoretical values for this quantity are overestimated. It is suggested that the long-range behaviour of the potential energy curves observed here for the first time represents the effective potentials including dominating resonance interactions and a possible slight contribution of the dispersion interactions as well as the R -dependent spin-orbit interaction.

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1. Introduction

The long-range interactions between two neutral atoms have received renewed attention over the past decade. This interest is stimulated first of all by the

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need for interpretation and explorations of several new phenomena occurring in an ensemble of ultracold atoms and molecules where the long-range interactions play a crucial role.

Interatomic potentials of diatomic systems at long distances have recently been determined with Le Roy and Bernstein method based on suitable extrapolation of experimental vibration energies beyond the range of the observed levels [1] or on fitting the experimental data to near dissociation expansion functions suggested in Ref. [2]. There is also a simpler and more direct method grounded on the analysis of spectral line wings profile broadened by interatomic interactions. Such a method is used in the present paper devoted to the study of the self-broadened resonance line of Zn at 213.8 nm ($4^1P_1 \leftarrow 4^1S_0$) which is one of the series of our papers concerning the electronic properties of the group IIb van der Waals dimers. Because the interaction in the upper molecular states 11_u and $^10_u^+$ is here dominated by resonance forces, the results obtained for the potential parameters give immediately the dipole transition moment or the oscillator strength for the considered molecular transitions. Particularly interesting seems to be the transition $^10_u^+ \leftarrow X$ occurring at long distances because it can be used for formation of molecules of the vibration energy near dissociation limit via photoassociation of free Zn atoms.

It should be mentioned that the self-broadening of the Zn 213.8 nm line has been studied years ago [3], however, either that paper or the recent studies of vibronic spectra associated with the upper states 11_u [4] and $^10_u^+$ [5, 6] performed with laser spectroscopy methods have not brought information concerning the long-range behaviour of the interaction potentials involved.

Additional motivation for our studies was a fact that the existing data, both experimental and theoretical, concerning an oscillator strength for the atomic transition $4^1P_1 \leftarrow 4^1S_0$ show considerable scatter of values [7].

2. Theoretical background

The line wings profile is analysed employing the well known classical approximation of the quasimolecular theory of the line shape broadened by interatomic interactions [8]. In the case of not very far wings produced during collisions at long distances where the interaction potential curves are of the form $V(R) = C_n R^{-n}$ this theory leads to an analytical formula for a normalized line shape function known as the Kuhn–Margenau profile [9, 10]:

$$I(\Delta\tilde{\nu}) = \frac{4\pi}{n} N_p \left| \frac{\Delta C_n}{hc} \right|^{3/n} |\Delta\tilde{\nu}|^{-(1+3/n)} \exp \left[-\frac{V_i(R(\Delta\tilde{\nu}))}{kT} \right], \quad (1)$$

where N_p is the perturbing atoms number density, $\Delta\tilde{\nu} = \tilde{\nu}_0 - \tilde{\nu}$, with $\tilde{\nu}_0$ being the line centre frequency (here in cm^{-1}), and $\Delta C_n = C_n^{(f)} - C_n^{(i)}$ is the difference of the potential parameters in the initial (i) and final (f) electronic states involved.

For a resonance line the difference potential at long distances, in general, is given by

$$\Delta V = \frac{C_3}{R^3} - \frac{\Delta C_6}{R^6}, \quad (2)$$

where C_3 is the resonant interaction constant in the excited state and ΔC_6 is a difference of the second-order van der Waals interaction constants in the excited and the ground states.

It can be expected that for sufficiently large R the van der Waals contribution in Eq. (2) is negligible and the Boltzmann factor in Eq. (1) can be omitted. Thus Eq. (1) simplifies to

$$I(\Delta\tilde{\nu}) = \frac{4\pi}{3} N_p \frac{C_3}{hc} |\Delta\tilde{\nu}|^{-2} = \frac{4\pi}{3} N_p \tilde{C}_3 |\Delta\tilde{\nu}|^{-2}. \quad (3)$$

Here and hereafter any tilde-assigned quantity dealing with energy is expressed in cm^{-1} .

The resonance interaction potential is easy to obtain on the level of the first-order perturbation theory of the interatomic interaction. The interaction constant C_3 depends on the orientation of the angular momentum J as following:

$$C_{3M} = \kappa C'_3, \quad (4)$$

where $\kappa = 1, -2$ for $M = 1, 0$, respectively, and C'_3 is given by

$$C'_3 = \frac{e^2 \hbar}{4\pi m c} \frac{f}{\tilde{\nu}_0} \quad \text{or} \quad \tilde{C}'_3 = \frac{e^2}{8\pi^2 m c^2} \frac{f}{\tilde{\nu}_0}. \quad (5)$$

Here e and m are the electron charge and mass, c is the speed of light, and f is the oscillator strength for the considered electronic transition.

Thus, the line shape formula for a single emitter surrounded by N_p perturbers can be rewritten as

$$I(\Delta\tilde{\nu}) = \frac{1}{6\pi} N_p \frac{e^2}{m c^2} \kappa \frac{f}{\tilde{\nu}_0} |\Delta\tilde{\nu}|^{-2} \quad (6)$$

and for N emitters $I_N(\Delta\tilde{\nu}) = N I(\Delta\tilde{\nu})$.

In the case of absorption the above consideration concerns the absorption coefficient spectrum $k(\Delta\tilde{\nu})$ through the relation

$$k(\Delta\tilde{\nu}) = \frac{\pi e^2}{m c^2} f g_{\text{if}} I_N(\Delta\tilde{\nu}), \quad (7)$$

where g_{if} is the statistical factor; for the molecular transition $\Omega \leftarrow \Sigma(0)$, $g_{\text{if}} = g_\Omega/g_e$ with g_Ω and g_e being the molecular and atomic statistical weights in the upper state. Such a designation of molecular states means that the Hund case (c) is assumed for a coupling scheme of different angular momentum in the molecule [11] which seems to be justified at long distances for the considered electronic states of Zn_2 . Thus, using Eq. (6) for $N_p = N$ one obtains

$$k(\Delta\tilde{\nu}) = \frac{1}{6} \left(\frac{e^2}{m c^2} \right)^2 \kappa \frac{g_\Omega}{g_e} \frac{1}{\tilde{\nu}_0} N^2 f^2 |\Delta\tilde{\nu}|^{-2} = A N^2 |\Delta\tilde{\nu}|^{-2}, \quad (8a)$$

where $A = \frac{1}{6} \left(\frac{e^2}{mc^2} \right)^2 \kappa \frac{g_\Omega}{g_e} \frac{1}{\tilde{\nu}_0} N^2 f^2$, or

$$k(\Delta\tilde{\nu}) = \frac{32\pi^4}{3} \kappa \frac{g_\Omega}{g_e} \tilde{\nu}_0 N^2 \tilde{C}'_3{}^2 |\Delta\tilde{\nu}|^{-2} = AN^2 |\Delta\tilde{\nu}|^{-2}, \quad (8b)$$

where $A = \frac{32\pi^4}{3} \kappa \frac{g_\Omega}{g_e} \tilde{\nu}_0 N^2 \tilde{C}'_3{}^2$.

On the basis of analogy to the similar electronic transitions for Hg₂ and Cd₂ it is expected that the blue wing of the considered line arises from the $1_u \leftarrow X$ transition where $g_\Omega/g_e = 2/3$ while the red one mainly from the $0_u \leftarrow X$ transitions with $g_\Omega/g_e = 1/3$, therefore $(g_\Omega/g_e)\kappa = 2/3$ for both wings. Thus it is seen that the resonance interaction leads to the Lorentzian profile centred on $\tilde{\nu}_0$ which in the wings of line is reduced to the form: $\text{const}/\tilde{\nu}_0^2$.

In the experimental studies it is convenient to consider the reduced absorption coefficient (in cm⁵) defined as follows:

$$k_r(\Delta\tilde{\nu}) = \frac{k(\Delta\tilde{\nu})}{N^2}. \quad (9)$$

This quantity is measured and analysed in this paper leading to the determination of the long-range behaviour of the potential energy curves and the oscillator strength f for the considered $^10_u, ^11_u \leftarrow X\Sigma_g(^10_g^+)$ transitions.

3. Experimental details

The experimental arrangement specified for absorption measurements is presented in Fig. 1. As a background source a super-quiet xenon lamp (Hamamatsu L2273) was used. The light from the lamp was filtered with a visible cut-off filter. This allows us to minimize the parasitic scattered light inside the spectrometer. The collimated light beam passed through a quartz cell 5 cm long and 3 cm diameter containing the zinc sample of high purity (6n). The cell was placed inside the oven with quartz windows. Temperature was measured by means of the calibrated Pt–PtRh thermocouple placed very close to the Zn deposit collected at the end of the side arm which was a bit longer than the cell. The transmission spectrum of the Zn vapour was recorded by the computer-controlled high resolution Rowland spectrophotometer [12]. Simultaneously a spectrum of the low pressure Cd lamp used as a reference for frequency calibration, was measured. The signals of the sample beam and the reference one were selected by a corresponding lock-in amplifier (SRS model SR810 DSP) and stored in a computer which also controlled the wavelength scanning activated by Camac system.

The stored data were recalculated to the reduced absorption coefficient as follows:

$$k_r(\Delta\tilde{\nu}) = \frac{k(\Delta\tilde{\nu})}{N^2} = \frac{\ln[I_0(\Delta\tilde{\nu})/I(\Delta\tilde{\nu})]}{N^2 d}, \quad (10)$$

where d is the length of the cell.

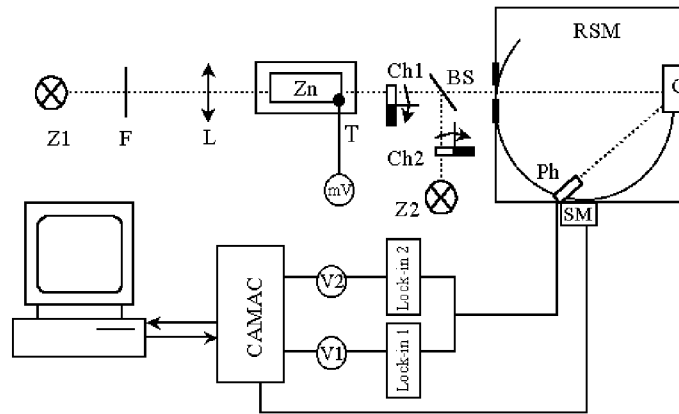


Fig. 1. The experimental setup. Z1 — Xe lamp, Z2 — Cd lamp, L — lens, F — filter, Ch1, Ch2 — choppers, T — Pt–PtRh thermocouple, BS — beam splitter, RSM — Rowland spectrometer, G — concave diffraction grating ($N = 174000$, $R = 6$ m), Ph — photomultiplier, SM — stepper motor, V1, V2 — digital voltmeters.

In the above formula $I_0(\Delta\tilde{\nu})$ and $I(\Delta\tilde{\nu})$ are the transmission spectra measured, respectively, at a room temperature (an empty cell) and for the cell at the temperature T determining the density of the saturated zinc vapour. Both spectra were corrected for a scattered light contribution and normalized to the same intensity outside the region of absorption in a sufficiently far red wing of the line.

The number density of Zn was calculated from the saturated vapour pressure as a function of temperature $p(T)$. Such data are usually taken from Nesmeyanov book [13], however, for Zn in the range of temperatures we are dealing with the Nesmeyanov data seemed to be insufficiently accurate. Therefore additional data of Hultgren et al. [14] were included to obtain the improved $p(T)$ function. Finally,

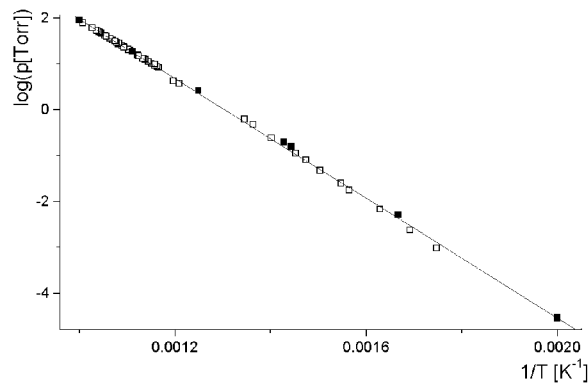


Fig. 2. Illustration of Eq. (11) for the saturated Zn vapour, \square data of Nesmeyanov, \blacksquare data of Hultgren et al., — fitted curve.

it was found that all of those data (for p in Torr) can be fitted by the following function presented in Fig. 2:

$$\log p = A \left(\frac{1}{T} \right) + B \quad (11)$$

with $A = -6523 \pm 24$, $B = 8.50 \pm 0.03$.

4. Results and discussion

Several series of measurements of the absorption spectrum around the 213.8 nm Zn line were performed for saturated Zn vapour at temperatures between about 475°C and 660°C, corresponding to the number densities $[\text{Zn}]$ between about 7.9×10^{15} and 3.4×10^{17} . At each temperature three runs of measurements were done, thus the final results were obtained on the basis of 50 scans.

Exemplary line profiles for the absorption coefficient are presented in Fig. 3. It is seen that they are symmetric and over a large frequency range they have the Lorentzian form: $k(\Delta\tilde{\nu}) = A\Delta\tilde{\nu}^{-2}$. We have observed such a behaviour for $|\Delta\tilde{\nu}| > 9 \text{ cm}^{-1}$ up to about 1000 cm^{-1} , but for evaluation of the C_3 or f value via inversion of Eqs. (8a,b) the region $10 < |\Delta\tilde{\nu}| < 400 \text{ cm}^{-1}$ characterized with the highest accuracy was considered.

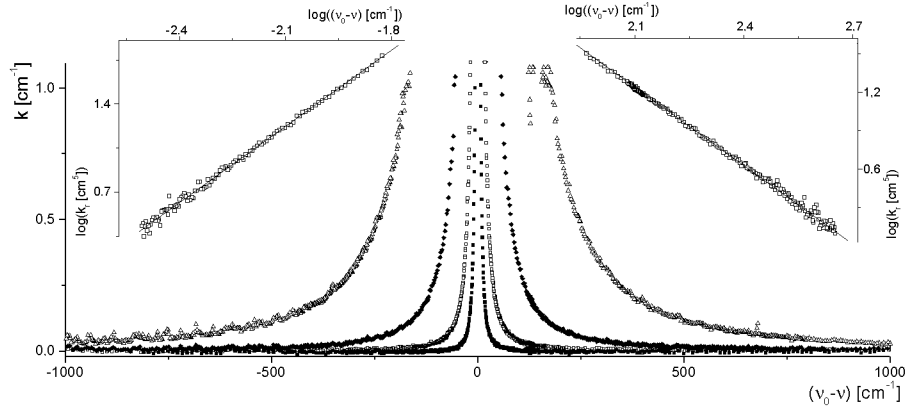


Fig. 3. Absorption profile of the Zn 213.8 nm line. From bottom up $[\text{Zn}] = 1.140 \times 10^{16}$; 3.261×10^{16} ; 1.069×10^{17} ; 3.500×10^{17} . In the latter case a trace of the 214.441 nm Cd reference line is seen at 127.5 cm^{-1} . Insets show the reduced absorption coefficient in the far blue (left) and red (right) wings of the line plotted on logarithmic scale. The experimental profiles and the linear fit of the slope (-2) are presented.

Experimentally determined profiles $k(\Delta\tilde{\nu})$ were corrected for the natural broadening which also leads to the Lorentzian line shape. Assuming that this effect and collisional one are statistically independent the observed profile $k(\Delta\tilde{\nu})_{\text{obs}}$ may

be considered as a convolution of the collisional profile $k(\Delta\tilde{\nu})_{\text{coll}}$ of the half width (FWHM) γ_{coll} and the natural profile $k(\Delta\tilde{\nu})_{\text{n}}$ of the half width γ_{n} . Hence the resulting half width is given by

$$\gamma_{\text{obs}} = \gamma_{\text{coll}} + \gamma_{\text{n}} = \gamma_{\text{coll}}(1 + \gamma_{\text{n}}/\gamma_{\text{coll}}). \quad (12)$$

A similar relation holds for relevant coefficients A being proportional to half widths. Then, the corrected values of A can be calculated as

$$A_{\text{corr}} = \frac{A_{\text{obs}}}{1 + \gamma_{\text{n}}/\gamma_{\text{coll}}}. \quad (13)$$

The effect of radiative contribution is significant near the line centre where collisional broadening is well described by the impact approximation which for resonance interactions yields

$$\gamma_{\text{coll}} = k_{JJ'} \frac{e^2 f}{8m_e c^2 \tilde{\nu}_0} N, \quad (14)$$

where $k_{JJ'}$ is a numerical constant depending only on the angular momentum of the ground and excited levels. Using Eq. (14) and the well-known expression for γ_{n} :

$$\gamma_{\text{n}} = \frac{4\pi e^2 \tilde{\nu}_0^2}{m_e c^2} \frac{g_i}{g_f} f, \quad (15)$$

one obtains

$$\frac{\gamma_{\text{n}}}{\gamma_{\text{coll}}} = \frac{32\pi \tilde{\nu}_0^3}{k_{JJ'} N} \frac{g_i}{g_f}. \quad (16)$$

The above correction for the considered ${}^1P_1 \leftarrow S_0$ transition of the frequency $\tilde{\nu}_0 = 46745.37 \text{ cm}^{-1}$ [15] and for $k_{JJ'} = 1.532$ following [16] amounts to $2.2344 \times 10^{15}/N$, so it is significant for $[\text{Zn}] < 10^{16} \text{ at/cm}^3$.

The coefficients A_{corr} obtained at different densities of vapour for the red and blue wings were used for determination of the C'_3 and f at different $\Delta\tilde{\nu}$ established for each case as the mean value of $\Delta\tilde{\nu}$ within the spectral range where the profile $k(\Delta\tilde{\nu})$ fulfils the relation $A\Delta\tilde{\nu}^{-2}$ with $A = \text{const}$. Since for such a case $\Delta\tilde{\nu}$ is univocally related to interatomic distance R , as final results the R -dependence of the oscillator strength for the transitions involved were deduced. These results are collected in Table I. It should be mentioned that under the conditions of this experiment (high temperatures) the Schumann–Runge bands of the O_2 molecule originating in $v'' = 1$ and even in $v'' = 2$ becomes observable disturbing significantly the blue wing profile. Therefore in Table I the data from the blue wing for $T > 600^\circ\text{C}$ are missed.

It is seen from this table that for $C'_3(\Delta\tilde{\nu})$ and $f(\Delta\tilde{\nu})$ a slight red asymmetry (stronger red wing) is observed. This effect reflects the fact that the blue and the red wing at the same $|\Delta\tilde{\nu}|$ arise from transitions occurring at different interatomic distances, being $\sqrt[3]{2}$ times larger for the red wing. This asymmetry disappears in the final results which show that f and C'_3 values obtained from the blue and

TABLE I

C'_3 and f values for increasing $\Delta\tilde{\nu}$ which in the fourth column are recalculated to R distances. At each T the first row includes data from the red wing and the second one — from the blue wing.

T	$\Delta\nu$ [cm ⁻¹]	C'_3 [eVÅ ⁶]	R [Å]	f
475.57	13.60	12.79 ± 0.1	24.75	1.351 ± 0.010
	12.58	12.48 ± 0.12	20.01	1.319 ± 0.007
490.08	15.90	12.82 ± 0.06	23.51	1.354 ± 0.007
	15.11	12.54 ± 0.05	18.85	1.325 ± 0.006
496.52	18.98	12.56 ± 0.04	22.02	1.327 ± 0.005
	17.19	12.18 ± 0.04	17.88	1.287 ± 0.004
504.92	22.40	12.48 ± 0.07	20.79	1.318 ± 0.007
	20.00	12.23 ± 0.04	17.02	1.292 ± 0.004
513.19	22.80	12.10 ± 0.02	20.46	1.279 ± 0.002
	22.13	11.96 ± 0.10	16.33	1.263 ± 0.011
521.19	34.76	12.19 ± 0.04	17.82	1.288 ± 0.004
	29.52	11.92 ± 0.06	14.82	1.259 ± 0.007
524.55	30.19	12.20 ± 0.03	18.68	1.288 ± 0.003
	27.32	11.99 ± 0.02	15.24	1.267 ± 0.002
535.31	36.33	12.06 ± 0.14	17.49	1.274 ± 0.005
	38.25	11.96 ± 0.05	13.61	1.263 ± 0.001
542.76	33.31	12.00 ± 0.02	17.98	1.268 ± 0.002
	40.84	11.76 ± 0.09	13.24	1.242 ± 0.009
549.28	57.24	12.09 ± 0.05	15.05	1.277 ± 0.005
	51.63	11.68 ± 0.08	12.22	1.234 ± 0.009
562.42	54.49	11.76 ± 0.05	15.16	1.243 ± 0.006
	51.48	11.57 ± 0.04	12.19	1.222 ± 0.004
567.47	68.69	11.73 ± 0.06	14.02	1.239 ± 0.006
	60.04	11.37 ± 0.01	11.52	1.201 ± 0.001
594.17	120.00	11.36 ± 0.03	11.51	1.200 ± 0.003
	109.18	11.07 ± 0.07	9.35	1.170 ± 0.007
609.55	150.93	11.29 ± 0.02	10.65	1.193 ± 0.002
	—	—	—	—
611.39	138.80	11.00 ± 0.04	10.85	1.162 ± 0.004
	—	—	—	—
627.08	177.52	10.93 ± 0.04	9.98	1.155 ± 0.004
	—	—	—	—
642.80	207.42	10.79 ± 0.05	9.43	1.140 ± 0.005
	—	—	—	—
660.18	233.59	10.58 ± 0.02	9.01	1.118 ± 0.002
	—	—	—	—

the red wing at a given R are the same within the error limits; however, they considerably decrease with R decreasing.

This behaviour cannot be explained by a temperature effect originating from the Boltzmann factor because the ground state potential in the region of R explored in this paper is very close to zero. The situation is illustrated in Fig. 4 which presents the long-range parts of the potential energy curves for the excited states 11_u and $^10_u^+$ derived in this work and for the ground state $X^10_g^+$. The latter is here represented by van der Waals potential C_6/R^6 where the C_6 constant amounting to $206 \text{ eV } \text{\AA}^6$ is calculated using Slater–Kirkwood formula as it has been done for Cd_2 in Ref. [17]. It was found that this long-range form for $R < 5 \text{ \AA}$ can be smoothly joined with the theoretical ground state potential for Zn_2 received in Ref. [18].

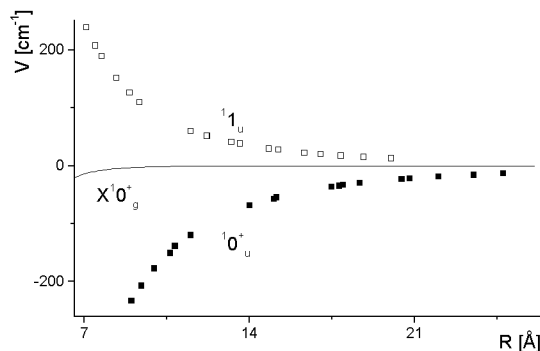


Fig. 4. The long-range potential energy curves for the excited states 11_u and $^10_u^+$ and for the ground state $X^10_g^+$, which is shifted up by 46745.37 cm^{-1} .

The other source of the observed R -dependence of the parameter C'_3 and f -value could be due to some distortion of the resonance interaction curves by influence of dispersion interactions in the excited states. Although the line profiles was found to be Lorentzian it can be expected that the dispersion interaction can contribute to the profile increasingly at larger spectral distances $\Delta\tilde{\nu}$. Therefore $C'_3(R)$ parameters should be thought as experimental parameters describing effective long-range interactions in the states 11_u and $^10_u^+$ of Zn_2 . They include the dominating resonance interaction and a possible rather slight contribution of dispersion interactions as well as the spin–orbit interaction which, in general, is also R -dependent. Moreover, the coupling scheme depends on interatomic distances going from the case (c) at long distance to the case (a) at distances where the electrostatic interatomic interactions are much stronger than the spin–orbit interaction in atoms. The present results cannot be compared with other data because this problem has not been studied yet so carefully for any of the group IIb dimers. However, some decrease in the molecular f -value in relation to the atomic value of f was observed for the resonance line of Cd at 228 nm [19] and Hg at 185 nm [20, 21].

It is interesting that the observed in this work R -dependence of C'_3 and f proved to be linear function of R^{-1} . In particular, Fig. 5 shows such results

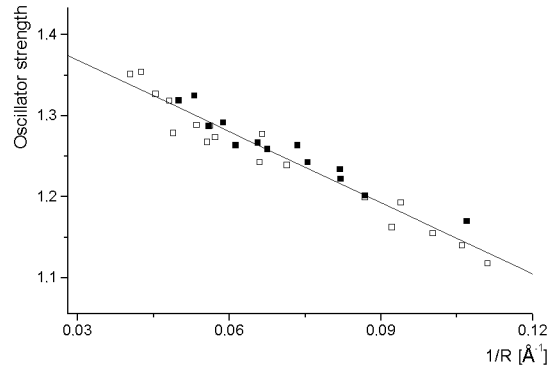


Fig. 5. The oscillator strength as a function of $1/R$ determined in this experiment; \square data from red wing, \blacksquare data from blue wing, — linear fit.

TABLE II
 f -values for the absorption $4^1S_0-4^1P_1$ transition in Zn.

Experimental	Theoretical
1.456 ± 0.010^a	1.49^e
1.61 ± 0.06^b	1.47^f
1.47 ± 0.03^c	
1.45 ± 0.15^d	

^athis work; ^bRef. [3]; ^cfrom the lifetimes, Refs. [22–27]; ^datomic beam absorption, Ref. [28]; ^enonrelativistic multiconfiguration Hartree–Fock, Ref. [29]; ^fCoulomb approximation, Ref. [30].

for the oscillator strength. The presented linear fit to the data extrapolated to $R \rightarrow \infty$ yields the oscillator strength $f_0 = 1.456 \pm 0.010$ for the Zn 213.8 nm line. In Table II this result is compared with the result of Ref. [3] obtained also from self-broadening of the same Zn line and with other experimental and theoretical data. From among a lot of such data we have chosen only those recommended in the review article by Doidge [7] as the most reliable. As it is seen the considerable disagreement exists between the f -value of Ref. [3] and the present result which being the most accurate agrees well with the remaining experimental data while the best theoretical results seem to be overestimated.

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