Long-range Interactions for the A^31_u and $B^30^+_u$ States of Cd₂ from Line Shape Measurements

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We report on very precise measurements of the absorption profile of the 326.1 nm resonance line of cadmium broadened by interactions with the ground state Cd atoms under thermal equilibrium. The experimental red wing profiles, corrected for the Boltzmann factor at different temperatures, are analysed in terms of the long-range potential parameters including dispersion and resonant interactions. The resonant interaction energy constant C_3 is calculated to be 2.31×10^{-2} eV Å³ and the van der Waals constant C_6 for difference potentials between the ground state $X^10_g^+$ and the excited states $B^30_u^+$ and A^31_u of Cd₂ derived experimentally are found to be (182.6 ± 8) eV Å⁶ and (289 ± 4) eV Å⁶, respectively. The former is determined for the first time and the latter is considerably corrected in comparison to earlier determination. The van der Waals constant C_6 for the ground state of Cd₂ is discussed and estimated to be $C_6^g = (247 \pm 40)$ eV Å⁶.

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1. Introduction

The long-range interactions between neutral atoms or molecules play an important role in a variety of physical situations, therefore they have been studied for a long time [1].

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The interest in accurate determination of interatomic potentials at large separations for various atom pairs has recently been enhanced because the knowledge of these data is necessary for interpretation of many new phenomena associated with collisions of cold atoms in the atoms traps as well as for stability prediction of such traps.

In the case of van der Waals dimers like Cd_2 a knowledge of the long-range portion of interaction potentials can be utilized for deriving these potentials at smaller separations when used an inversion procedure for far wings of the spectral line broadened by interaction with a collision partner. In our earlier paper [2] we have shown that such a method gives the interatomic potentials of a quite good quality over a wide range of separations for both electronic states involved. However, since it is based on the intensity information the accuracy of final results depends critically on a precision of the line profile measurements on the absolute intensity scale.

The present paper is devoted to determination of van der Waals constants for Cd₂ in the lowest excited states A and B correlated with the asymptote Cd(5³P₁)+Cd(5¹S₀). So far, there have appeared only a few papers dealing with spectroscopy of interatomic interactions for Cd₂. In particular, the profile of the self-broadened Cd(5³P₁-5¹S₀) $\lambda = 326.1$ nm line in absorption has been investigated in our paper [3] including estimates of the main parameters for a difference potential between the ground state $X^{10}_{\text{g}}^{+}$ and the excited state A^{31}_{u} of Cd₂. The similar study has been undertaken in [4], however, the precision of absolute intensity measurements achieved in that paper was not enough high in order to improve the earlier results, particularly those concerning the long-range behaviour of the potentials involved.

On the other hand, the $B^30^+_{\rm u} \leftarrow X^10^+_{\rm g}$ transition in Cd₂ has been explored in the molecular beam spectroscopy experiments [5, 6] yielding the vibrational frequency ω_0 and the anharmonic correction $\omega_0 x$ for the electronic states B and Xas well as the difference $r'_{\rm e} - r''_{\rm e}$ between the equilibrium positions in these states. Although such data give reliable information about the wells region, they, unfortunately, are insufficient for determining potential curves at large separations.

It should be noted that interaction potentials for a number of electronic states of the Cd_2 dimer have also been derived theoretically with different methods [7–9]. The best agreement with experiment show the predictions based on partly semi-empirical calculations developed by Czuchaj et al. [7], however, they do not cover the long-range region of interatomic distances.

Although long-range interatomic forces, being a well-defined theoretical problem [10], have been intensively studied in the past [11] and the quite accurate numerical results for dispersion coefficients, in particular for the dipole–dipole one (C_6) , are now available for a variety of simple species [11, 12], this is not a case for Cd and other many-electron atoms. The difficulty is that precise calculations of these coefficients require the knowledge of the line strengths and frequencies for dipole transitions between a given state and all the other states (including those in continuum) [13] or, alternatively, the dynamic dipole polarizability at the imaginary frequency for both interacting atoms [14, 15]. These characteristics for Cd_2 are not available at present either via purely quantum theory or in semi-empirical approach.

This situation motivated us to carry out new very precise measurements of the profile of the Cd 326.1 nm line with the primary aim to determine the long-range section of the potential energy curves for the above-mentioned molecular states. Some data have recently been briefly reported [16] and final results based on more comprehensive line shape analysis are presented in this paper.

2. Experiment

The absorption profile of the $5^3P_1 \leftarrow 5^1S_0$ (326.1 nm) resonance line of cadmium was measured with a 6 meter concave grating Rowland spectrometer. This instrument, described in details elsewhere [17], has recently been carefully realigned achieving, in a first order spectrum at about 320 nm, a limit resolution of 0.2 cm⁻¹ being appropriate for investigation of even very near wings of line.

A super-quiet xenon lamp (Hamamatsu L2273) was used as a background source. A high stability of this lamp enabled us to use a single beam configuration for absorption measurement while the second channel was utilized for a precise wavelength scaling with a spectrum of the low pressure Cd lamp.

In order to decrease the intensity of a parasitic scattered light inside a spectrometer, the light beam from the lamp was filtered with a visible cut-off filter. In this way the scattered light level was reduced to 0.5 percent of the continuum at about 326 nm. This residual effect was carefully measured and it was accounted for in the computer program which analysed the spectral data.

A pure cadmium vapour contained in a sealed quartz (suprasil) cell of 5 cm length was placed inside a 60 cm long oven closed with quartz windows. Additional insulating cover and carefully adjusted operating conditions for the heating system assured a fairly uniform temperature over the cell volume. We have worked with a saturated vapour, therefore the great pains were taken to control and accurate measure the cell temperature determining a vapour pressure. This was done with Pt-PtRh thermocouples calibrated by the State Bureau of Standards with accuracy of 0.4 K at the highest temperatures.

The measured quantity was the absorption coefficient of the vapour as a function of a frequency difference $\Delta \nu = \nu_0 - \nu$, where ν_0 is a frequency of the line center including a collisional shift. It was tested that in our case the frequency ν_0 exactly corresponds to the center frequency of the emission line from the Cd lamp used (see Fig. 1).

With a view to the main goal of the present work we have concentrated on a precise determination of the absolute absorption coefficient (in cm^5)

$$k_{\rm a}(\Delta\nu) = -\ln[I(\Delta\nu)/I_0(\Delta\nu)]/[{\rm Cd}]^2 d, \tag{1}$$



Fig. 1. Comparison of the transmission profile (\bullet) from the sample cell at low [Cd] with the emission profile (\circ) of the reference Cd lamp.

where [Cd] is the vapour number density and d denotes the optical path length. The most important was the red wing of the 326.1 nm Cd line for $\Delta \nu$ up to about 500 cm⁻¹. To obtain the line profile over such a range the measurements were carried out at several vapour densities between 6.07×10^{15} cm⁻³ ($T \simeq 633$ K) and 3.09×10^{18} cm⁻³ ($T \simeq 963$ K).

The values of [Cd] were calculated from the vapour pressure (p) chosen from experimental data recommended by Nesmeyanov [18] as the most reliable ones for liquid cadmium in the considered range of temperatures. We have found that those data for p in torr can be fitted by the following function:

$$\log(p) = A - B/T - C/T^2 + D/T^3$$
with $A = 7.0973$, $B = 913.6$, $C = 33145 \times 10^2$, and $D = 815649 \times 10^3$. (2)

3. The spectrum, its interpretation and analysis

The experimental profile of the self-broadened Cd 326.1 nm line resulting from several measurements at different Cd densities is presented in Fig. 2. A significant asymmetry of this profile proves that the resonant long-range interaction is here very weak in comparison with the dispersion interactions. However, for accurate analysis, particularly in the near red wing, the contribution arising from the resonant interaction is expected to have some meaning.



Fig. 2. The profile of the self-broadened Cd $(5^1S_0 - 5^3P_1)$ 326.1 nm line presented as the absolute absorption coefficient $k(\Delta\nu)$ from several measurements at different number densities of Cd (in cm⁻³). Open circles: 6.07×10^{15} ; closed circles: 7.7×10^{16} ; open diamonds: 3.12×10^{17} ; closed diamonds: 9.3×10^{17} .



Fig. 3. Red wing of the Cd 326.1 nm line in a logarithmic scale from several scans at various vapour densities (open and closed symbols).

Figure 3 shows an example of the red wing profile from several experimental runs at vapour densities appropriate for observation of this wing over a wide frequency range. Let us note that this spectrum is very like to those observed for the red wing of the 326.1 nm line perturbed by heavy rare gases (see e.g. [4]) and for other van der Waals systems such as metal IIb-rare gas diatomics. In particular, at the frequency $\Delta \nu$ of about 10 cm⁻¹ in Fig. 3 a slight shoulder appears which on the analogy of similar spectral features occurring for the above-mentioned systems may be interpreted as a rainbow satellite [19] associated with the existence of a very shallow minimum (of several cm^{-1}) of the potential difference function, here $\Delta V^0 = V(B^30^+_{\mu}) - V(X^10^+_{\sigma})$, at long interatomic distances. The observation of this satellite provides important and quite new information about a long-range behaviour of the potential energy curve of the state B of Cd_2 . As it is seen this state plays a crucial role in formation of the near red wing of the Cd 326.1 nm line contrary to an existing opinion that the state B contributes to only the blue wing of this line. On the other hand, it is rather well established that the potential difference $\Delta V^1 = V(A^3 1_u) - V(X^1 0_g^+)$ is a monotonic function of R and it gives rise to the red wing profile over a whole spectral range including a very far wing.

The line shape analysis in terms of the interaction potentials parameters is based on the quasimolecular treatment of the optical collision [19]. We use the well-known (see e.g. [2] and references therein) classical approximation of this theory yielding the so-called quasistatic profile, which holds in the wings of line except for such regions where satellite bands appear. The method is especially simple in the case of not very far wings produced during optical collision at long distances, where the interaction potential curves can be described by the monotonic function of the inverse power form

$$V(R) = C_n R^{-n}.$$
(3)

In such a case the quasistatic one-perturber line wing profile, specified for the absorption $e \leftarrow g$ dipole-allowed transition in the absorber-perturber diatomic system, or the cross-section for such an effect (in cm²) is given by

$$k_n(\Delta\nu) = C f_0 N \frac{4\pi}{n} \frac{g_\Omega}{g_e} \left| \frac{\Delta C_n}{hc} \right|^{3/n} |\Delta\nu|^{-(n+3)/n} \exp\{-V_g[R(\Delta\nu)]/k_BT\}.$$
(4)

Here $C = \pi e^2/m_e c = 0.88528 \times 10^{-12}$ cm, where e and m_e are the electron charge and mass and c is the light speed; f_0 is the oscillator strength of the unperturbed transition, N is the perturber density, g_{Ω} and g_e are the statistical weights of the excited molecular and parent atomic state and ΔC_n is a difference of the C_n coefficients in the upper and lower electronic states involved. The Boltzmann factor $\exp\{-V_g[R(\Delta\nu)]/k_BT\}$ describes the population of the ground state as a function of $\Delta\nu$ related to the difference potential $\Delta V(R)$ by means of the classical Franck–Condon principle as follows:

$$\Delta \nu = \Delta V(R)/hc. \tag{5}$$

Equation (4) allows us to determine, from the experimental line wing profile, the coefficient ΔC_n and the index *n* in the potential formula (3) if the Boltzmann factor as a function of $\Delta \nu$ over the relevant spectral range is known. In the near red wing this function is very close to be equal to 1 and Eq. (4) can be then simplified to

$$k_n(\Delta\nu) = A_n N \Delta\nu^{-s} \tag{6}$$

with $A_n = Cf_0(4\pi/n)(g_\Omega/g_e)|\Delta C_n/hc|^{3/n}$ and s = 1 + 3/n. The above equation in a double-logarithmic scale represents a straight line with a slope equal to -s. In particular, for a van der Waals (n = 6) interaction s = 1.5 and this case is known as a Kuhn profile which usually is utilized for determination of ΔC_6 coefficients.

To return to Fig. 3, it is seen that the red wing of the 326.1 nm Cd line can be interpreted in the frame of such a model over a wide range of the frequency $\Delta\nu$ excluding vicinity of the weak satellite band. However, before any analysis it is necessary to have a spectrum free of the Boltzmann factor which can be determined from the temperature dependence of this spectrum. In the case of the 326.1 nm Cd line this problem was studied in Ref. [20] where, in particular, the $V_{\rm g}(\Delta\nu)$ function in the red wing was derived. Using that result (presented in Fig. 4) the Boltzmann factor functions at suitable temperatures were calculated and experimental profiles determined in the present work were divided by them.



Fig. 4. The ground state potential plotted against $\Delta \nu$ from Ref. [20] in the red wing of the Cd self-broadened line at 326.1 nm.

The profiles free of the Boltzmann factor were analysed in terms of the long-range potential parameters including van der Waals and resonant interactions. We have observed the influence of the latter as a slight increase in the slope coefficient above 1.5. In order to account for this effect quantitatively, a knowledge of a resonance interaction constant C_3 is needed. Unfortunately, there are no experimental results for this constant, therefore we have it calculated employing the literature data on the oscillator strength for the $5^3P_1-5^1S_0$ transition in Cd.

As it is well known, the resonant interaction potential has the following form:

$$V(R) = \gamma_m C_3 R^{-3} \tag{7}$$

with $\gamma_m = +1$ and -2 for the 1_u and 0_u states, respectively, and $C_3 = \frac{C}{8\pi^3} \frac{f_0}{\nu_0}$. Thus, the quasistatic profile arising from the resonant interaction, according to Eq. (6) is

$$k_3(\Delta\nu) = A_3 N \Delta\nu^{-2},\tag{8}$$

where $A_3 = C^2 (3\pi^2 g_e hc)^{-1} f_0^2 \nu_0 = 88.23089 \times 10^{-28} f_0^2 / \nu_0$ cm². Hence, for the considered case, taking $f_0 = 0.0016$ [21] and $\nu_0 = 30656.13$ cm⁻¹ [22] we obtain $C_3 = 2.31 \times 10^{-2}$ eV Å³ and $A_3 = 73.68 \times 10^{-38}$ cm³.

It is noteworthy that $|\gamma_m| \times g_{\Omega}$ is equal to 2 for both the molecular states $A^3 1_u$ and $B^3 0^+_u$, therefore although the interaction energy on the red wing is twice as large as on the blue wing, the resonance profile is symmetrical.

Finally, the experimental absorption coefficient defined by Eq. (1) in the red wing of line, when corrected for the Boltzmann factor, was considered as normalized to the Cd vapour density the quasistatic profile given by

$$k(\Delta\nu) = k_6(\Delta\nu)/N + k_3(\Delta\nu)/N = A_6\Delta\nu^{-1.5} + A_3\Delta\nu^{-2}$$
(9)

with A_3 presented above. This function could be fitted with accuracy of 0.1% to the experimental profiles in two regions of $\Delta \nu$: between 3–6 cm⁻¹ and for $\Delta \nu > 20$ cm⁻¹ yielding the A_6 coefficient which was the only fitting parameter.

The exemplary results of such an operation are presented in Figs. 5 and 6. The former demonstrates also an influence of the Boltzmann factor on the spectrum. Let us note that the second component of Eq. (9), in the case considered, only a bit changes the general shape of the fitted profiles which in a double-logarithmic scale remain the straight lines, however, their slopes are slightly greater than -1.5. It is particularly visible in the near wing, where the effective slope parameter s reaches a value of about -1.54.

As mentioned previously, the near red wing profile is produced during the transitions to both the excited states A^31_u and $B^30^+_u$ so, ignoring a collisional coupling between these states at long distances, the coefficient A_6 is

$$A_6^{(1)} = C' \left(\frac{2}{3}\sqrt{\Delta C_6^1} + \frac{1}{3}\sqrt{\Delta C_6^0}\right),\tag{10}$$

while, in the far wing only the state $A^3 1_u$ is involved, thus

$$A_6^{(2)} = \frac{2}{3} C' \sqrt{\Delta C_6^1},\tag{11}$$

where $C' = \frac{2\pi}{3\sqrt{hc}}Cf_0$.



Fig. 5. The far red wing of the Cd 326.1 nm line. (o) the direct experimental data; (•) the same data corrected for a Boltzmann factor using $V_{\rm g}(\Delta\nu)$ from Fig. 4. The continuous line is a fit of the function given by Eq. (9).



Fig. 6. The near red wing of the Cd 326.1 nm line for two separated scans at $[Cd] = 6.07 \times 10^{15} \text{ cm}^{-3}$. The continuous line is a fit of the function given by Eq. (9).

4. Results and discussion

The mean values for coefficients A_6 from several experimental line wing profiles are found to be $A_6^{(1)} = (422 \pm 1.5) \times 10^{-38} \text{ cm}^{3.5}$ and $A_6^{(2)} = (302 \pm 2.0) \times 10^{-38} \text{ cm}^{3.5}$, yielding the van der Waals constants: $\Delta C_6^0 = (182.6 \pm 8) \text{ eV} \text{ Å}^6$ and $\Delta C_6^1 = (289 \pm 4) \text{ eV} \text{ Å}^6$. The error bounds of the van der Waals constants result directly from the uncertainties in the mean A_6 values which are due to scattering data obtained from different measurements. As mentioned, for a given experimental profile the uncertainty of the A_6 coefficient was the rms error of the fit amounting to not more than 0.1%. In addition to the presented uncertainties the ΔC_6 may be affected by systematic errors due to possible deviations from a real values of the oscillator strength f_0 and the Cd number densities, which determine the scale factor for the intensity of the spectrum. In this paper we have chosen the best available information concerning these data but final results can be easily revisited if any better data appear.

It should be noted that the essential role for accurate determination of van der Waals constants from line shape analysis plays an interpretation of the spectrum in terms of the long-range interaction forces. In the present paper we accounted for the dispersion and resonance interactions, however, our experimental spectra over almost the same frequency regions can be fitted by a pure Kuhn profile, ignoring the $k_3(\Delta\nu)$ component in Eq. (9), as it has been done in Refs. [3, 4]. In such a case for $A_6^{(1)}$ and $A_6^{(2)}$ one obtains, respectively, 466×10^{-38} and 316.6×10^{-38} that yields the van der Waals constants (in eV Å⁶) $\Delta C_6^1 = 318.5 \pm 4$ and $\Delta C_6^0 =$ 284 ± 10 . It is seen that these results differ very much from the values corrected for resonance interaction although the neglecting of this interaction might seem to be justified.

Only on a very limited scope our results can be compared with other data. The coefficient ΔC_6^0 is derived here for the first time and only the ΔC_6^1 has been considered previously. In particular, ΔC_6^1 has been estimated to be 168 eV Å⁶ in our earlier paper [3] and that result was adopted in Ref. [4]. Our present determination deviates considerably from the previous result, however, the performance of the measurements as well as the procedure of the data analysis have been improved significantly since then, so we believe that the present result is much closer to the real one.

In order to obtain C_6 constants for the excited states A and B we need to know this constant for the ground state. At present it can be only estimated using some approximate formulae. We have chosen the Slater-Kirkwood (SK) [23] and the London (L) [24] formulae which, as it was shown by Tang et al. [11] and by other authors (e.g. [25, 26]), give the upper and lower bounds to the C_6 constant. In the case of the homonuclear diatomics, for C_6 in a.u., these formulae can be written as follows:

$$C_6(SK) = \frac{3}{4}\alpha \sqrt{\alpha N_{\text{eff}}},\tag{12}$$

$$C_6(\mathbf{L}) = \frac{3}{4} \alpha^2 \gamma_{\text{eff}}.$$
(13)

Here α is the static dipole polarizability, N_{eff} is the effective number of electrons giving rise to the dispersion dipole–dipole interaction or, more exactly, the number N satisfying the oscillator strength sum rule, and γ_{eff} is the effective transition energy estimating the sum over all transitions[†].

The $N_{\rm eff}$ number for Cd₂ has been obtained from the empirical formula derived in Ref. [27] (Eq. (6)), which gives $N_{\rm eff} = 3.7569$, and $\gamma_{\rm eff} = 0.25061$ we have accepted after Ref. [28]. Regarding the dipole polarizability for Cd in the ground state, the data available are scattered within the limits about $(40-50)a_0^3$ ([28–30] and references therein). Values close to the lower limit have been obtained from measurements of the static polarizability α while the higher values — from an extrapolation of the dynamic polarizability curve $\alpha(\omega)$ to $\omega = 0$. There exist also theoretical results from self-consistent field (SCF) calculations, however they are not enough accurate [30].

We calculated $C_6(SK)$ and $C_6(L)$ for $\alpha = 40.49a_0^3$, according to Miller and Bederson [29], and for $\alpha = 49.65a_0^3$ obtained by Goebel and Hohm [28] from measurements of the frequency dependence of the refractive index of cadmium vapour. The results are collected in Table. Assuming that Eqs. (12) and (13) still give the upper and lower bounds for C_6 it seems that a mean value of $C_6(SK)$ and $C_6(L)$ estimates reasonably the real value of C_6 . The problem concerns, however, a value of the dipole polarizability which needs to be established more precisely in future studies.

TABLE

Values of C_6 [in eV Å⁶] for the ground state Cd atom pair obtained from Eqs. (12) and (13) at different polarizabilities α .

$\alpha (a_0^3/\text{\AA}^3)$	$C_6(SK)$	$C_6(L)$	Average
40.49/6.0	223.8	184.1	204
49.65/7.357	303.9	276.86	290

Tentatively we estimate $C_6^g = (247 \pm 40) \text{ eV } \text{\AA}^6$ for the ground state which yields $C_6^1 = (536 \pm 44) \text{ eV } \text{\AA}^6$ and $C_6^0 = (430 \pm 48) \text{ eV } \text{\AA}^6$ for the excited states A^31_u and $B^30_u^+$, respectively. We emphasise that uncertainty of this determination is due to uncertainty of the van der Waals constant for the ground state of Cd₂.

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[†]In the original formulation the number of the outer shell electrons is used for $N_{\rm eff}$ and the ionization energy is used for $\gamma_{\rm eff}$.

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