Eur. Phys. J. D **23**, 217–222 (2003)
DOI: 10.1140/epjd/e2003-00035-7

THE EUROPEAN PHYSICAL JOURNAL D

Laser-induced fluorescence study of the influence of N_2 and CH_4 on the ^{114}Cd intercombination line

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Received 29 November 2001 / Received in final form 12 July 2002

Abstract. Using a laser-induced fluorescence method a detailed analysis of profiles of the ^{114}Cd 326.1 nm line perturbed by N_2 and CH_4 was performed which revealed deviations from the ordinary Voigt profile. These deviations are shown to be consistent with fits of experimental profiles to an asymmetric Voigt profile. Coefficients of the pressure broadening, shift and collision-time asymmetry are determined and compared with those calculated for van der Waals interaction potential.

PACS. 32.70.-n Intensities and shapes of atomic spectral lines – 33.70.-w Intensities and shapes of molecular spectral lines and bands – 34.20.-b Interatomic and intermolecular potentials and forces, potential energy surfaces for collisions

1 Introduction

In recent papers [1–4] precise studies of pressure broadening and shift of the cadmium 326.1 nm (5^1S_0–5^3P_1) intercombination line perturbed by noble gases (Ar, Kr, Xe) and two non-polar molecular gases (H_2 and D_2) performed by means of a laser-induced fluorescence (LIF) method were reported. In these studies the line shapes were analysed in terms of a speed-dependent asymmetric Voigt profile [5] and the role of the correlation between the collisional- and Doppler-broadening as well as of the finite duration of collisions were thoroughly investigated. These effects were found to be particularly important in the cases of perturbation by heavy, *i.e.* high-polarizability, noble gas atoms (Ar, Kr, Xe) [1–3]. In these cases a line shape analysis revealed departures from the ordinary (symmetric) Voigt profile which, however, were shown to be consistent with fits of experimental line shapes to speed-dependent asymmetric Voigt profiles. An important feature is that the asymmetry of the 326.1 nm Cd line produced by Ar, Kr and Xe is towards the red, *i.e.* in the same direction as the pressure shift of this line caused by these three noble gases. This agrees with theoretical predictions for the van der Waals potential that a red shift should be associated with a higher intensity red wing [6,7]. On the other hand, in the case of perturbation by molecular gases H_2 and D_2 [4] in the pressure range up to 30 torr the shapes of the 326.1 nm line were found to be symmetric and could be fitted very well to the ordinary Voigt profile.

In the present work we extend our previous measurements of the shapes of the 326.1 nm Cd line using nitrogen and methane as perturbing gases. It seems to be interesting to compare the Cd-noble-gas atoms as well as Cd–H_2 and Cd–D_2 broadening and shift measurements with those obtained in the case of Cd–N_2 and Cd–CH_4 since N_2 and CH_4 (like H_2 and D_2) have no permanent electric dipole moment. Special attention in the present study was focused on the problem of asymmetry of the 326.1 nm Cd line in order to establish whether the correlation effects and the effect of finite duration of collisions have any influence on the resulting profile.

The only experimental study [8] of the influence of N_2 on the Cd intercombination line dealt with the pressure shift of this line and involved measurements at a fixed pressure of 1050 torr and temperature 1670 K. However, no experimental data on the shape and the width of the 326.1 nm line perturbed by N_2 were reported in reference [8]. To our knowledge, in the present investigation the broadening, shift and asymmetry in the Cd–N_2 and Cd–CH_4 mixtures have been systematically investigated for the first time.

2 Experimental setup

The experimental procedure and the details of the line shape analysis used in the present work were identical to those described in earlier papers from this laboratory in which the LIF technique was applied to study the pressure broadening, shift and asymmetry of the Cd 326.1 nm line [1–4]. In order to avoid the hyperfine and isotopic structure of the line the ^{114}Cd isotope was used. The fluorescence cells containing cadmium were filled with nitrogen or methane and cut-off from the vacuum system

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and then mounted in an oven at a temperature of 450 K with a stability up to 1 K. The perturbing gas pressure was varied between 5 and 300 torr for N_2 and between 2 and 350 torr for CH_4 at room temperature. This corresponds to the density of perturbing gas between 6×10^{16} and 1.2×10^{19} molecules/cm^3, whereas the density of cadmium in the cell was kept constant 4×10^{12} atoms/cm^3. Thus in the present experiment the Cd–Cd interactions do not play any role.

Profiles of the intercombination cadmium line were registered using a digital laser spectrometer described elsewhere [4]. An actively stabilized single-frequency Coherent CR 899-21 ring dye laser equipped with an intracavity frequency doubler CR 8500, operating on DCM dye was pumped by an INNOVA-400 argon-ion laser. The ring laser provided single mode UV output continuously tunable over 60 GHz. The line width of this laser line was about 1 MHz. The intensity of the fluorescence signal was measured by a thermoelectrically cooled photomultiplier working in the photon counting mode. Frequency calibration of the ring laser was performed using its fundamental (red) line directed to a confocal Fabry-Perot interferometer (FPI) with a free spectral range of 1.5 GHz and to a 100 cm long iodine cell operated at a temperature of 35 °C. The FPI transmission peaks and I_2 absorption spectrum were recorded simultaneously with the fluorescence signal for frequency calibration. Because the incident UV laser beam was linearly polarized in the vertical direction the collection optics arm contained a linear polarizer set at the "magic angle" (rotated 54.7° from the vertical), so the collection optics system was insensitive to effects due to anisotropy of fluorescence [5,9]. The photon counting process was performed by an electronic system built in the CAMAC standard described elsewhere [10]. All the data: fluorescence signal, UV laser output power, FPI transmission peaks and I_2 absorption spectrum were stored on a PC computer for further evaluation. Measurements were performed at the laser power between 10 and 140 μW. Tests have shown that for the laser power up to 160 μW the power-broadening has negligible effect on the line shape.

3 Data analysis

Figure 1 shows profiles of the ^{114}Cd line perturbed by nitrogen at pressures 5, 154, and 343 torr. In this pressure region the maxima of the line located at the wave numbers $\widetilde{\nu}_m$ were found to be shifted towards lower wave numbers (red shift) with respect to the unperturbed wave number $\widetilde{\nu}_0$. Simulations analogous to those performed in our previous work [1] showed that for Cd–CH_4 as well as for Cd–N_2 at pressure between 2 and 350 torr the Dicke-narrowing and correlation between dephasing and velocity-changing collisions make negligible contributions to the profiles of the 326.1 nm line. This means that the Doppler and collisional broadening can be in this case approximately treated as statistically independent effects. The resultant line profile $I(\widetilde{\nu})$ can then be written as a convolution of the Gaussian distribution corresponding to the Doppler temperature T of the gas and the profile $I_C(\widetilde{\nu})$ describing the collisionally-broadened component of the total profile.

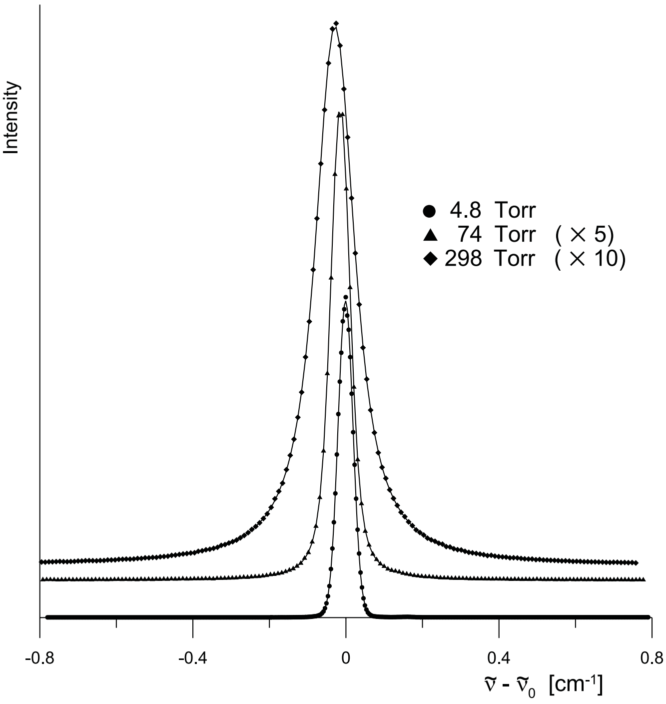

Fig. 1. Profiles of the ^{114} Cd line perturbed by nitrogen at pressures 4.8, 74, and 298 torr.

In order to take into account the effects caused by the finite duration of collisions in the present analysis we assumed the profile $I_C(\widetilde{\nu})$ to be represented as a sum of a Lorentzian and a dispersion profile [6,11]

$$I_C(\widetilde{\nu}) = \frac{1}{\pi} \frac{(\gamma_L/2) + \chi(\widetilde{\nu} - \widetilde{\nu}_0 - \Delta)}{(\widetilde{\nu} - \widetilde{\nu}_0 - \Delta)^2 + (\gamma_L/2)^2}, \qquad (1)$$

where χ is the collision-time asymmetry parameter. Here γ_L is the Lorentzian width and $\Delta = \widetilde{\nu}_m - \widetilde{\nu}_0$ is the shift of the line with respect to $\widetilde{\nu}_0$.

In the semiclassical approximation the Lorentzian width and the shift can be evaluated from the well-known formula of the Baranger impact theory [7,12] in which both γ_L and Δ are expressed in terms of the products of the diagonal elements of the scattering matrix $\hat{S} = \hat{U}(-\infty, \infty)$ for the initial and final states of the radiating atom suitably averaged over angular coordinates, impact parameter ρ and the Maxwellian distribution $f(\mathbf{v})$ of relative velocities $\mathbf{v}$ of the colliding particles (see *e.g.* Eq. (4) in Ref. [3]). Here $\hat{U}(-\infty, \infty)$ denotes the time-evolution operator $\hat{U}(t_1, t_2)$ over the time interval between $t_1 = -\infty$ and $t_2 = \infty$ for a system consisting of a radiating atom and perturbing molecule. Ciuryło *et al.* [13,14] have shown that the collision-time asymmetry parameter χ depends on the products of the elements of the time-evolution operators $\hat{U}(-\infty, t_0)$ or $\hat{U}(t_0, \infty)$, where t_0 denotes the time of closest approach. The parameter χ can then be presented as an average of such products over

t_0 and, as usual, over impact parameters, angular coordinates and the Maxwellian distribution of velocities (*cf.* Eq. (5) in Ref. [3] or Eq. (16) in Ref. [13]).

In the classical limit time ordering in the $\hat{U}(t_1, t_2)$ operators is ignored and these operators are replaced by the exponentials of the phase-shift functions

$$\eta(t_1, t_2) = \frac{1}{\hbar} \int_{t_1}^{t_2} \Delta V(r(t)) \mathrm{d}t, \tag{2}$$

where $\Delta V(r) = V_\mathrm{u}(r) - V_\mathrm{l}(r)$ is the difference of adiabatic interaction potentials for the upper (u) and lower (l) levels of the emitting atom, respectively, expressed as a function of the distance $r(t)$ between emitter and the perturber at time t.

The width (γ_L) and shift (Δ) parameters describing the Lorentzian component of the profile depend now on the exponentials of the $\eta(-\infty, \infty)$ functions. For a van der Waals difference potential $\Delta V(r) = -\Delta C_6\ r^{-6}$ with $\Delta C_6 = C_6^\mathrm{u} - C_6^\mathrm{l}$ being the difference of force-constants for the upper and lower states, respectively, γ_L and Δ are then given by the well-known formulae of the classical Lindholm-Foley impact theory (see Eqs. (271, 272) in Ref. [7]).

Following Ciuryło *et al.* [13] the collision-time asymmetry parameter χ in the classical limit can be written in the form

$$\chi = 2\pi N \,\mathrm{Im} \int \mathrm{d}^3\mathbf{v}\ f(\mathbf{v})\ v \int_0^{+\infty} \rho\ \mathrm{d}\rho \times \int_{-\infty}^{+\infty} \mathrm{d}t_0 \Big\{ 1 + \exp\big[-\mathrm{i}\eta(\infty, -\infty)\big] - \exp\big[-\mathrm{i}\eta(t_0, -\infty)\big] - \exp\big[-\mathrm{i}\eta(\infty, t_0)\big] \Big\}. \tag{3}$$

For the van der Waals potential this formula yields results close to those obtained by Traving [15] on the basis of the Anderson-Talman theory (*cf.* [7]) and by Al-Saquabi and Peach [16]. In their analysis of the non-Lorentzian behaviour of spectral line shapes, Walkup *et al.* [17,18] have shown that for attractive van der Waals potential ($\Delta C_6 > 0$), the collision time asymmetry can be expressed in the following form:

$$\chi = -\frac{\gamma_\mathrm{L}}{2} a |\Delta C_6|^{1/5} v^{-6/5}, \tag{4}$$

where a is a numerical factor whose value depends on the definition of the relative speed v. Assuming v to be equal to the rms relative velocity $v = v_\mathrm{rms} = (3kT/\mu)^{1/2}$ with μ being the reduced mass Walkup *et al.* [18] obtained $a = 0.547$.

Following Harris *et al.* [5] the profile $I(\tilde{\nu})$ calculated from the convolution of the Gaussian distribution and $I_\mathrm{C}(\tilde{\nu})$ given by equation (1) will be referred to as the asymmetric Voigt profile $I_\mathrm{AVP}(\tilde{\nu})$. For $\chi = 0$ the profile becomes identical with the ordinary Voigt profile $I_\mathrm{VP}(\tilde{\nu})$.

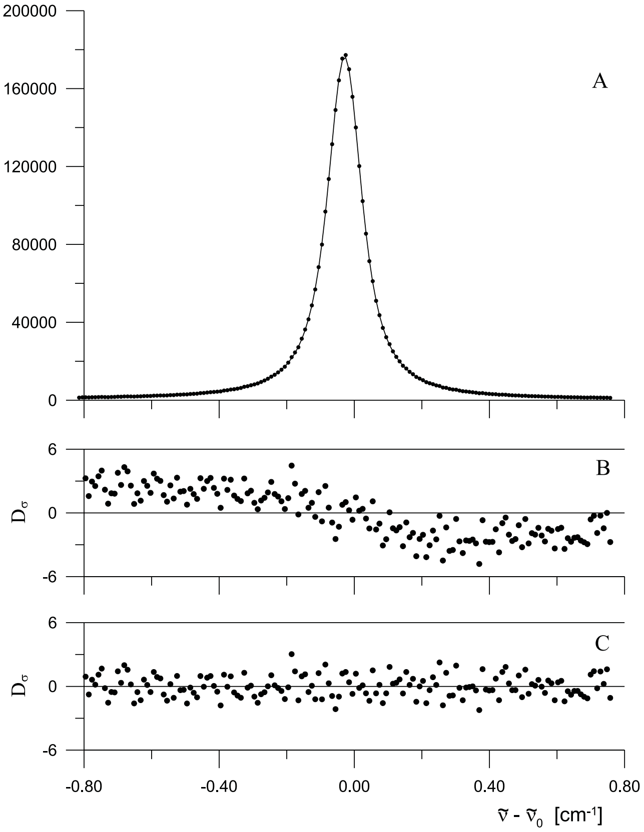

Fig. 2. The shape of the Cd 326.1 nm line perturbed by N_2 at a pressure of 298 torr; (A) experimental points together with the fitted AVP profile (full curve), (B) and (C) weighted residuals D_σ between experimental and fitted VP and AVP profiles, respectively.

In order to examine the influence of the collision time asymmetry on the total line shape we fitted our experimental data both to a Voigt profile $I_\mathrm{VP}(\tilde{\nu})$ and an asymmetric Voigt profile $I_\mathrm{AVP}(\tilde{\nu})$. The best fit procedure was performed using the Marquardt algorithm [19] for a least-squares estimation of nonlinear parameters. In the case of Voigt profile $I_\mathrm{VP}(\tilde{\nu})$ the fitting parameters were: the Gaussian width γ_D, the Lorentzian width γ_L, the line shift Δ and constant background signal. For the asymmetric Voigt profile $I_\mathrm{AVP}(\tilde{\nu})$ we also fitted the asymmetry parameter χ.

Figure 2A shows an example of the shape of the 326.1 nm ^{114}Cd line perturbed by N_2 at a pressure of 298 torr at room temperature. The fitted $I_\mathrm{AVP}(\tilde{\nu})$ profile is plotted as the solid line. In order to examine the quality of the fit we used the weighted residuals $D_\sigma(\tilde{\nu})$ between the experimental $I_\mathrm{exp}(\tilde{\nu})$ and fitted $I_\mathrm{fit}(\tilde{\nu})$ profiles:

$$D_\sigma(\tilde{\nu}) = \frac{I_\mathrm{exp}(\tilde{\nu}) - I_\mathrm{fit}(\tilde{\nu})}{u_c(\tilde{\nu})}. \tag{5}$$

where $u_c(\tilde{\nu})$ is the uncertainty of the measured profile at wave number $\tilde{\nu}$.

In Figure 2B we plotted the residuals $D_\sigma(\tilde{\nu})$ for the case when $I_\mathrm{fit}(\tilde{\nu})$ was given by the Voigt Profile $I_\mathrm{VP}(\tilde{\nu})$. We can see the systematic departures from zero in the

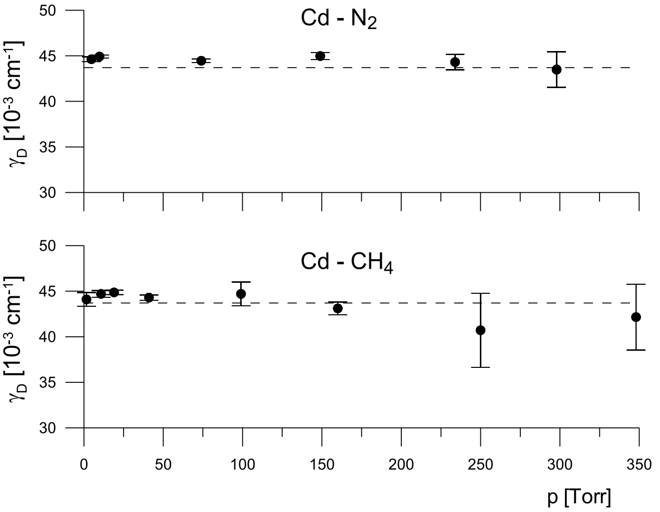

Fig. 3. Plots of the Doppler halfwidths γ_D of the Cd 326.1 nm line against the pressure of N_2 and CH_4. Dashed line – theoretical Doppler halfwidth corresponding to the cell temperature (450 K). Error bars indicate the values of standard deviations.

line core as well as on line wings that can be regarded as a manifestation of the line asymmetry. Figure 2C shows the residuals $D_\sigma(\widetilde{\nu})$ for the case when for the fit the asymmetric Voigt profile $I_{AVP}(\widetilde{\nu})$ was used. In this case the values of residuals are spread uniformly around zero which confirms the quality of the fit. The same behaviour was observed for Cd–CH_4. We can thus conclude that in the case of the Cd–N_2 and Cd–CH_4 systems the effect of finite duration of collisions has a noticeable influence on the profile of the Cd 326.1 nm line.

4 Results and discussion

Figure 3 shows the experimental values of the Doppler widths γ_D of the ^{114}Cd 326.1 nm line perturbed by N_2 and CH_4 determined from the fit of the asymmetric Voigt profile to the experimental data plotted as a function of pressure. It is evident that there is no dependence of the Doppler width on the perturbing gas pressure. It confirms the statistical independence at Doppler and pressure broadening for the Cd–CH_4 and the Cd–N_2 systems. The average Doppler widths γ_D were found to be $(44.1 \pm 1.5) \times 10^{-3}$ cm^{-1} and $(43.6 \pm 2.2) \times 10^{-3}$ cm^{-1} for the Cd–N_2 and the Cd–CH_4 systems, respectively. These values agree with a theoretical Doppler width 43.7×10^{-3} cm^{-1} corresponding to the cell temperature (450 K).

The Lorentzian widths γ_L and shifts Δ determined from the fit of the asymmetric Voigt profile to our experimental data are plotted in Figure 4 as functions of the perturbing gas density N. As can be seen both the Lorentzian width γ_L and shift Δ are linearly dependent on N. The experimental values for the pressure broadening $\beta = \gamma_L/N$ and shift $\delta = \Delta/N$ coefficients, determined from the slopes of the straight lines shown in Figure 4 are listed in Table 1. These are the first measurements of the pressure broadening and shift coefficients for the Cd–N_2 and Cd–CH_4 systems.

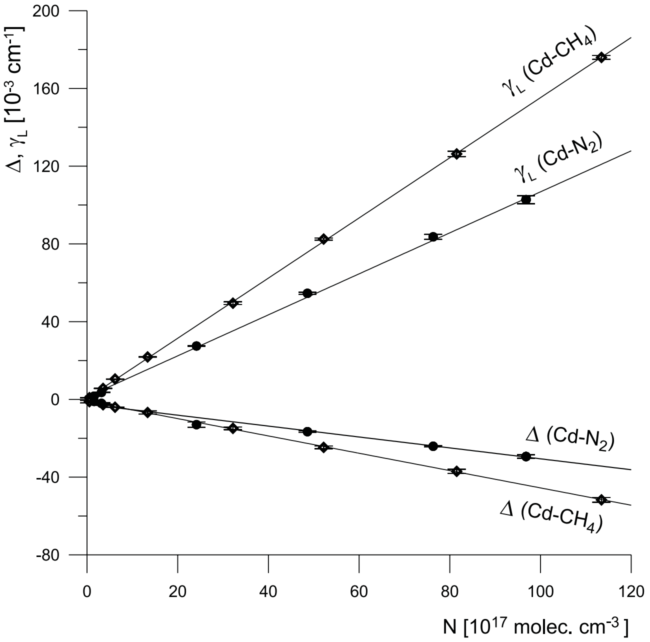

Fig. 4. Plot of the Lorentzian halfwidth γ_L and shift Δ of the Cd 326.1 nm line against the density N of N_2 and CH_4. Error bars indicate the values of standard deviations.

Table 1. Comparison of experimental values for the β, δ (in units 10^{-20} cm^{-1}/molecules cm^{-3}) and κ (in units 10^{-21}/molecules cm^{-3}) coefficients with those calculated for the van der Waals (v.d.W.) interaction potential. For experimental data standard deviations are given as well.

Cd–N_2	β	δ	κ
Experimental values	1.054(9)	–0.281(7)	–0.36(8)
Theoretical (v.d.W.), Eq. (3)	0.933	–0.338	–0.60
Theoretical (v.d.W.), Eq. (4)	–	–	–0.45
Cd–CH_4	β	δ	κ
Experimental values	1.548(5)	–0.446(4)	–0.81(7)
Theoretical (v.d.W.), Eq. (3)	1.255	–0.455	–0.66
Theoretical (v.d.W.), Eq. (4)	–	–	–0.50

In their measurements of the pressure shift of the 326.1 nm Cd line perturbed by N_2 Greenstein *et al.* [8] have determined the value $\Delta = -0.085 \pm 0.009$ cm^{-1} for the nitrogen density $N = 6.1 \times 10^{18}$cm^{-3} at a temperature 1670 K. Unfortunately, the value of the pressure shift coefficient δ has not been reported in reference [8]. Assuming a linear dependence of Δ on N in their experiment we can roughly estimate δ to be about -1.4×10^{-20}cm^{-1}/molecules cm^{-3}. This value is about 5 times larger than the value determined in the present work. Let us note, however, that our measurements were performed at the cell temperature 450 K which is about 3.7 times lower than that in reference [8].

Figure 5 shows the values for the collision-time asymmetry χ for Cd–N_2 and Cd–CH_4 determined from the fit

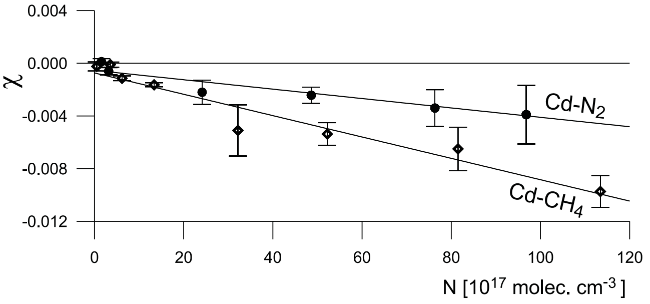

Fig. 5. Plot of the asymmetry parameter χ of the Cd 326.1 nm line against the density N of N_2 and CH_4. Error bars indicate the values of standard deviations.

of the asymmetric Voigt profile to the experimental profiles plotted against N_2 and CH_4 density. The straight lines were fitted to the experimental points and the values for the asymmetry coefficient $\kappa = \chi/N$ were determined and listed in Table 1. These are the first measurements of the collision-time asymmetry coefficient for the Cd–N_2 and Cd–CH_4 systems.

Let us note that both for Cd–N_2 and Cd–CH_4 the experimental values of the pressure shift coefficient as well as the collision-time asymmetry coefficient were found to be negative ($\delta < 0$, $\kappa < 0$). This means that for the 326.1 nm Cd line perturbed by N_2 and CH_4 a red shift is associated with a higher intensity at the red wing of the line. Similar behaviour was observed earlier in our laboratory for the Cd 326.1 nm line perturbed by heavier noble gases (Ar, Kr, Xe) [1–3] as well as by the Lewis group [20,21] for the Ca 422.7 nm line perturbed by Ar, Kr and Xe. Negative values of both the shift and collision-time asymmetry coefficient were also found by Romalis *et al.* [22] for the rubidium D_1 and D_2 lines perturbed by N_2. We should note, however, that in the case of perturbation by He and Ne the Lewis group [20,21] and Romalis *et al.* [22] found positive collision-time asymmetry coefficients for the Ca 422.7 nm and Rb D_1 and D_2 lines, respectively. This means that in these cases the red shift is associated with a higher intensity at the blue wings of these lines.

In order to interpret the pressure broadening, shift and collision-time asymmetry data the realistic interaction potentials for both the excited (5^3P_1)- and ground (5^1S_0)-state Cd atoms colliding with N_2 or CH_4 molecules should first be known. Unfortunately, up to now for Cd–N_2 and Cd–CH_4 such potentials, or – more precisely – *ab initio* potential surfaces are not available. For this reason in the present study we have attempted to interpret our experimental results assuming the Cd–N_2 and Cd–CH_4 interaction potentials in the form of a van der Waals potential. We are aware that this represents a very crude picture of the processes under investigation, but nevertheless we believe that it can be used as a first step to get more insight into collision dynamics for such systems.

The van der Waals C_6 force-constants depend on the quantum numbers of the energy levels of the Cd atom and the polarizability α of the perturbing molecule. The quantum-mechanical calculation of C_6 requires the knowledge of line strengths for all transitions in the radiating atom which involve the upper and lower states [23]. Unfortunately, the line strengths for many transitions involving the 5^1S_0 and 5^3P_1 states are not known yet. Therefore, in order to estimate C_6 we used here an approximate formula given by Unsöld [24]: $C_6 = \alpha e^2 \left\langle r^2 \right\rangle$ where e is the elementary charge and $\left\langle r^2 \right\rangle$ is the quantum-mechanical expectation value of r^2 in a given state of the radiating atom. The values of $\left\langle r^2 \right\rangle$ were calculated using the Coulomb approximation. Here we used the experimental values $\alpha = 1.76$ Å^3 [25] and $\alpha = 2.59$ Å^3 [26] for the polarizability of the N_2 and CH_4 and obtained the following values for the C_6 constants (in atomic units): for Cd–N_2 76.7 and 126 for the cadmium 5^1S_0 and 5^3P_1 states, respectively and for Cd–CH_4: 113 and 185 for the cadmium 5^1S_0 and 5^3P_1 states, respectively. Substituting these constants into equation (2) and performing numerical integration of equation (3) we obtained for the asymmetry coefficient $\kappa = \chi/N$ the value $\kappa = -0.60 \times 10^{-21}$/molecules cm^{-3} for Cd–N_2 and $\kappa = -0.66 \times 10^{-21}$/molecules cm^{-3} for Cd–CH_4. On the other hand, the calculation based on equation (4) with $a = 0.547$ as derived by Walkup *et al.* [18] yields $\kappa = -0.45 \times 10^{-21}$/molecules cm^{-3} for Cd–N_2 and $\kappa = -0.50 \times 10^{-21}$/molecules cm^{-3} for Cd–CH_4.

The comparison with the data given in Table 1 shows that the agreement between experimental values of κ and the theoretical ones computed on the basis of a van der Waals potential is poor, but nevertheless the theoretical values of κ are of the same order of magnitude as the experimental ones. Using the same values of C_6 constants we have also calculated the pressure broadening (β) and shift (δ) coefficients using the well-known formulae of the classical Lindholm-Foley impact theory (Eqs. (271, 272) in Ref. [7]). Results of such calculations are given in Table 1.

As it is seen, both for Cd–N_2 and Cd–CH_4 the theoretical values of β and δ obtained for the van der Waals potential are in astonishingly good agreement with experimental data. All experimental results reported in the present work were determined at a fixed cell temperature 450 K. No detailed study of the influence of temperature on the line shape was made. However, for Cd–N_2 we have performed additional measurements at temperature 724 K and density 90.3×10^{17} molecules/cm^3. We found that the ratios of the Lorentzian widths γ_L and the shifts Δ at the temperatures 450 K and 724 K are $\gamma_L(450)/\gamma_L(724) = 0.898$ and $\Delta(450)/\Delta(724) = 0.828$. This values agree well with theoretical value for these ratios equal to 0.867 resulting from the Lindholm-Foley theory for the van der Waals potential which predicts both for γ_L and Δ the $T^{0.3}$ dependence. The experimental value of the asymmetry parameter χ at 724 K was found to be -0.0045 with standard uncertainty 0.0042. This means that the precision of the asymmetry determination at 724 K is too small to deduce any quantitative conclusion about temperature dependence.

In the conclusion, we should emphasize that the simple adiabatic Libdholm-Foley theory which represents the classical limit of the quantum-mechanical Baranger [12] theory has usually been applied to cases of perturbation by noble-gas atom. Thus it might well be expected that the theory which treats the noble-gas atoms as polarizable spheres could also be applicable to perturbations due to molecules. Obviously a van der Waals potential alone does not represent a sufficient approximation to the real potentials in the Cd–N_2 and Cd–CH_4 systems. Furthermore, in calculations of β, δ and κ coefficients one should take into account not only the distance r between Cd atom and perturbing molecules but also the spatial orientations of perturbers. Concluding, we hope that the results of the present experiment may encourage theoreticians to search for realistic potentials for Cd–N_2 and Cd–CH_4 systems.

This work was supported by the grant No. 5 P03B 066 20 (354/P03/2001/20) from the Committee for Scientific Research. The authors wish to thank the Referees for critical remarks regarding the first version of this paper.

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