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Energy pooling to the Ba 6s6p $^1P_1^0$ level arising from collisions between pairs of metastable Ba 6s5d ^3D_J atoms

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Abstract. The energy-pooling rate coefficient for the Ba(6s5d ^3D_J) + Ba(6s5d ^3D_J) → Ba(6s^2 $^1S_0$)+ Ba(6s6p $^1P_1^0$) process has been measured. The barium atoms were excited by a cw diode laser tuned to the frequency of the 791.3 nm intercombination line and the metastable atoms in the 6s5d ^3D_J state were produced due to radiative and collisional depopulation of the laser-excited 6s6p $^3P_1^0$ state. The measurements were performed at ^{138}Ba number densities of about 4×10^{10} cm^{-3} and at 30 mbar argon as the buffer gas. Most of the barium ground state atoms in the excitation zone were transferred to the triplet metastable state at the laser pump power applied. The energy pooling rate coefficient was determined by comparing the fluorescence intensity of the barium 553.6 nm resonance line and the fluorescence intensity of the intercombination line 791.3 nm. In addition, the populations of the metastable atoms were probed with low intensity laser radiation from a single mode ring dye laser. The rate coefficient was found to be $k = 1 \times 10^{-10}$ cm^3 s^{-1} at $T = 730$ K.

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1 Introduction

The lowest-lying barium excited states 6s5d $^1D_2$ and 6s5d ^3D_J are metastable states with extremely long radiative lifetimes [1] due to the fact that their radiative relaxation to the ground state is dipole forbidden, and in addition, they are of the same parity as the barium ground 6s^2 $^1S_0$ state. The barium metastable states can be populated by optical excitation of higher-lying states and their subsequent radiative and collisional relaxation. Collisionally enhanced population of the metastable levels can be very efficient. For instance, as shown in [2], a very large fraction of the barium ground-state atoms can be transferred into the metastable states when the 6s6p $^3P_1^0$ state is optically excited with relatively low power laser radiation and the barium atoms collide with noble gas atoms. Recently, the detailed studies on the population and depopulation processes related to collisions between barium metastables and noble gas atoms were reported [3–5].

The energy pooling process occurs in collisions between two excited atoms, where one highly excited atom and one ground state atom is produced. Since the high number density of the barium atoms excited to the metastable states can be achieved, very pronounced energy pooling effects can appear in barium vapours. These effects were observed in barium for the first time in the experiments dealing with laser enhanced ionisation spectroscopy. Ion signals were obtained in barium vapours by optical excitation of low-lying states. Energy pooling was assumed to be the major mechanism for producing the barium ions [6]. It was found that strong energy pooling effects produce significant non-linearities in the spectra, which have to be taken into account in quantitative spectroscopy.

Recently, a quantitative study on energy pooling processes involving barium atoms excited to the 6s6p $^3P_1^0$, 6s5d $^1D_2$ and 6s5d ^3D_J states has been reported by Neuman *et al.* [7]. Barium atoms in higher lying P, D and F states were produced by the energy pooling collisions. Contributions by several pairs of the initial states are possible for most of the investigated energy pooling channels. These contributions were not resolved but upper limits for rate coefficients for assumed particular combinations of initial states have been determined.

Here we present the measurements of the rate coefficient for an energy pooling process in barium, which involves the triplet metastable states as the initial states and 6s6p $^1P_1^0$ as the final state:

$$\begin{aligned} &\mathrm{Ba}(6s5d\ ^3D_J) + \mathrm{Ba}(6s5d\ ^3D_J) \\ &\quad \rightarrow \mathrm{Ba}(6s6p\ ^1P_1^0) + \mathrm{Ba}(6s^2\ ^1S_0) + \Delta E. \end{aligned} \tag{1}$$

The investigated process is interesting and instructive for

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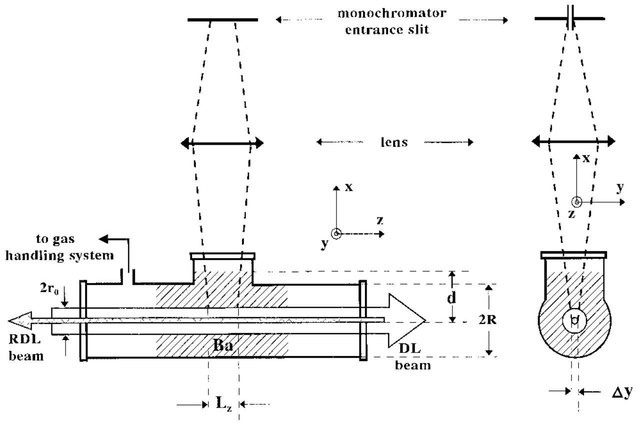

Fig. 1. Experimental arrangement; DL: diode laser, RDL: ring dye laser, d: length of the optical path through the layer of non-excited barium vapour, L_z: length of the observed fluorescence slab defined by the height of the monochromator entrance slit and imaging ratio (1:2), Δy: width of the observed fluorescence slab defined by the monochromator entrance slit width, $2r_0$: diameter of the pump laser beam, $2R$: heat-pipe diameter.

several reasons. The final state is the barium resonance state, and, regarding the energy defects, this process is expected to be the most probable energy pooling process when two triplet barium metastables collide. The energies of the 6s5d $^3\mathrm{D}_J$ sublevels are 9034 cm^{-1}, 9215 cm^{-1} and 9596 cm^{-1} for $J = 1,\ 2,$ and 3, respectively. The energy of the barium 6s6p $^1\mathrm{P}_1^0$ resonance state is 18060 cm^{-1}, and for particular $^3\mathrm{D}_J + {}^3\mathrm{D}_{J'}$ pairs, the process (1) is characterised by energy defects ΔE ranging from $+8$ cm^{-1} to $+1132$ cm^{-1}. Certainly, the most interesting case is almost resonant $^3\mathrm{D}_1 + {}^3\mathrm{D}_1(\Delta E = +8\ \mathrm{cm}^{-1})$ collision. However, in the present experiment, the triplet metastable sublevels were completely mixed due to collisions with noble gas atoms, and we were not able to resolve the contributions of each particular pair. Only an upper limit of the rate coefficient for the particular $^3\mathrm{D}_J + {}^3\mathrm{D}_{J'}$ collision could be derived. The obtained value for the energy pooling rate coefficient is an average value at given experimental conditions.

The average energy defect ΔE for the process (1) at experimental temperature is $\sim kT$ and positive. Since the energy defects for other higher-lying barium states are significantly larger than kT and negative, one can expect that other energy pooling effects involving two barium triplet metastables are less probable than the process (1). As shown in [2,3], it is easy to obtain a large population in the triplet states *via* laser excitation of the 6s6p $^3\mathrm{P}_1^0$ state in a noble gas atmosphere. Thus, under specific experimental conditions, the process (1) can appear as the most pronounced energy pooling process in barium vapour.

Among other energy pooling processes, Neuman *et al.* [7] investigated this particular process also. As reported in [7], the fluorescence of the resonance line at 553.6 nm arising from the 6s6p $^1\mathrm{P}_1^0 \rightarrow 6\mathrm{s}^2\ {}^1\mathrm{S}_0$ transition is severely trapped and only the lower limit for the rate coefficient for the process (1) could be obtained. In the present paper we report on an experiment in which the resonance radiation in the excitation zone was not trapped. On the basis of the investigations reported in [3–5] we have determined the optimal experimental conditions and defined the experimental procedure which allowed us to evaluate the rate coefficient for the energy pooling process under consideration.

2 Experiment

The experimental arrangement, shown schematically in Figure 1, is the same as was used in our previous investigations on barium [3,5]. Therefore, it will be described only briefly. The barium metal was contained in a three-arm stainless-steel cell (diameter 3 cm) filled with argon as the buffer gas. The cell was resistively heated and the generated barium vapour was optically excited by a pump laser tuned to the frequency of the 791.3 nm Ba intercombination line. In fact, the frequency of the pump laser was adjusted to the centre of the $6\mathrm{s}^2\ {}^1\mathrm{S}_0 \rightarrow$ 6s6p $^3\mathrm{P}_1^0$ transition of the ^{138}Ba isotope (71.7% natural abundance). The pump laser was a single-mode frequency stabilised cw Hitachi laser diode (maximum power: 10 mW, linewidth: less than 20 MHz). The widened beam of the pump laser had the diameter $2r_0 = 6$ mm and nearly homogeneous power distribution in the barium vapour. In order to measure the metastable populations, the counter-propagating probe beam of a single mode Spectra Physics ring dye-laser was shone through the zone excited by the diode laser. The probe beam had the diameter of about 1 mm and its power was reduced to 5 μW. In order to measure the population densities of the collisionally populated Ba metastable levels, the probe laser was tuned to the wavelengths of either 582.8 nm or 602.1 nm. The respective probe transitions are 6s5d $^3\mathrm{D}_1 \rightarrow$ 5d6p $^3\mathrm{P}_J^0$ and 6s5d $^1\mathrm{D}_2 \rightarrow$ 5d6p $^1\mathrm{P}_1^0$. The absorption of both the pump and probe beam was measured by photodiodes. The fluorescence of the barium intercombination and resonance line was observed through the cell side-arm at right angles to the laser beam propagation direction. The central part of the fluorescence zone parallel with its axis (slab width: $\Delta y \approx 1$ mm, slab length $L_z \approx 6$ mm) was imaged to the entrance slit (slit width: 500 μm, slit height: 3 mm) of a 1 m McPherson monochromator and the fluorescence signals were detected by a RCA S-20 photomultiplier.

3 Model and method

3.1 Rate equations

If the Ba 6s6p $^3\mathrm{P}_1^0$ state is optically excited, the population and depopulation processes due to collisions between excited barium atoms and noble gas atoms can be described within a five level model [3] which is schematically illustrated in Figure 2. The relevant collisional and radiative transition rates are denoted by R_{ij} and A_{ij}, respectively, where $i,\ j = \mathrm{g, p, s, t}$ and r. The subscripts

Table 1. Barium radiative transition probabilities and the cross sections for the excitation transfer processes in barium induced by collisions with argon.

$A_{\rm rg}(10^8\ {\rm s}^{-1})$	$A_{\rm pg}(10^5\ {\rm s}^{-1})$	$A_{\rm pt}(10^5\ {\rm s}^{-1})$	$\sigma_{\rm ps}(10^{-16}\ {\rm cm}^2)$	$\sigma_{\rm st}(10^{-16}\ {\rm cm}^2)$
$1.19^{\rm a}$	$5.3^{\rm b}$	$3^{\rm b}$	$5.7^{\rm c}$ $4.6^{\rm d}$	$0.037^{\rm c}$ $0.015^{\rm d}$ $0.021^{\rm e}$

$^{\rm a}$: [8]; $^{\rm b}$: [9]; $^{\rm c}$: [4]; $^{\rm d}$: [3]; $^{\rm e}$: [5].

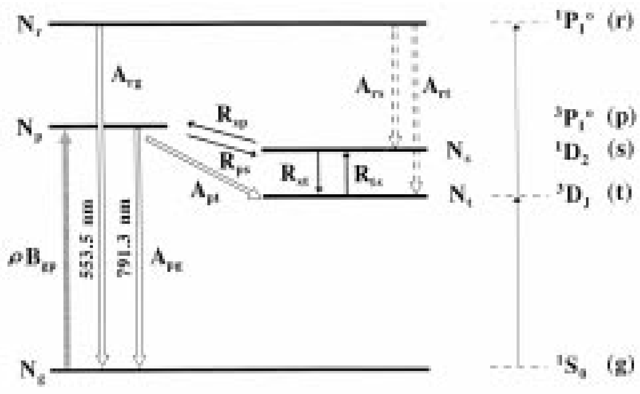

Fig. 2. Partial term diagram of barium including the relevant radiative and collisional transitions (for the explanation of the labels see the text).

g, p, s, t and r label the ground state 6s^2 $^1S_0$, 6s6p $^3P_1^0$ state, singlet metastable state 6s5d $^1D_2$, triplet metastable state 6s5d ^3D_J and the resonance state 6s6p $^1P_1^0$, henceforth referred to as $^1S_0$, $^3P_1^0$, $^1D_2$, ^3D_J and $^1P_1^0$, respectively. The radiative rates A_{ij} and the cross-section values σ_{ij} corresponding to the rates R_{ij} for collisions with argon, are listed in Table 1. The collisional rates are defined as $R_{ij} = \sigma_{ij}\ v^{\rm rel}_{\rm Ba-Ar} N_{\rm Ar}$, where $v^{\rm rel}_{\rm Ba-Ar}$ is the mean Maxwellian relative velocity and $N_{\rm Ar}$ is the argon number density.

The main collisional depopulation channel for the $^3P_1^0$ state is the $^3P_1^0 \to {}^1D_2$ transition, while both collisional mixing among the 6s6p $^3P_J^0$ sublevels and the $^3P_1^0 \to {}^3D_J$ collisional transitions are negligible in comparison with the $^3P_1^0 \to {}^1D_2$ transition [3,4]. As for the ^3D_J states, they are populated radiatively and collisionally *via* the $^3P_1^0 \to {}^3D_J$ transitions and collisionally *via* the $^1D_2 \to {}^3D_J$ transitions. At noble gas pressures above 2 mbar, complete intramultiplet mixing among the 6s5d ^3D_J substates occurs [3,4]. As shown in reference [3], the quenching processes for Ba metastable levels are negligible in comparison with other rates.

If the rate equations for the number densities N_i of the excited barium atoms are considered, the diffusion of metastable atoms out of the laser excitation zone should be taken into account too. The radiative lifetimes of the metastable barium states are very long, and the losses caused by diffusion of the barium metastable atoms out of the excitation zone depend strongly on noble gas pressure [3,7]. The corresponding average value of the diffusion removal rate for the whole excitation volume can be estimated as $\overline{W}_D \approx D/r_0^2$, where D is the diffusion coefficient and r_0 is the radius of the cylindrical excitation volume.

Presuming that the experimental conditions have been attained, in which the relevance of the involved processes just coincide with the scheme described above (see Fig. 2), the steady-state rate equations for the p, s and t levels in the excitation zone can be written as:

$$\frac{{\rm d}N_{\rm p}}{{\rm d}t} = 0 = -(A_{\rm pg} + A_{\rm pt} + R_{\rm ps})N_{\rm p} + \rho B_{\rm gp} N_{\rm g} + R_{\rm sp} N_{\rm s} \tag{2}$$

$$\frac{{\rm d}N_{\rm s}}{{\rm d}t} = 0 = -(R_{\rm st} + R_{\rm sp} + W_D^{\rm s})N_{\rm s} + R_{\rm ps} N_{\rm p} + R_{\rm ts} N_{\rm t} \tag{3}$$

$$\frac{{\rm d}N_{\rm t}}{{\rm d}t} = 0 = -(R_{\rm ts} + W_D^{\rm t})N_{\rm t} + A_{\rm pt} N_{\rm p} + R_{\rm st} N_{\rm s}. \tag{4}$$

Here, $\rho B_{\rm gp}$ is the laser pump rate for the optical transition g → p ($B_{\rm gp}$: Einstein coefficient for the absorption, ρ: spectral power density).

In principle, equation (4) should include a term associated with the energy pooling to the resonance state. However, for the typical values of the triplet number density ($N_{\rm t} \approx 5 \times 10^{10}\ {\rm cm}^{-3}$), the depopulation of the triplet metastable levels due to energy pooling is negligible in comparison with other effects. This can be proved by taking into account the estimated values for the energy pooling rate coefficients (about $10^{-11}\ {\rm cm}^3\ {\rm s}^{-1}$) in barium [7]. Therefore, the rate equation (4) does not include this term. The population of both metastable states through the radiative channels (labelled with radiative rates $A_{\rm rs}$ and $A_{\rm rt}$ in Figure 2) from the resonant state is negligible because of the low number density of the $^1P_1^0$ atoms. Accordingly, the corresponding terms are omitted in equations (3) and (4). It has to be noted that the population densities N_i in equations (2), (3) and (4) are dependent on the space co-ordinates. Their spatial distributions are governed by the shape of the pump laser power distribution, diffusion of the triplet metastables out of the excitation zone and collisional mixing among the metastable states and the optically pumped state. The position dependence of N_i in the rate equations (2), (3) and (4) is given *via* W_D terms, which are functions of the distance from the beam axis. To determine the position dependence of the exited populations the appropriate diffusion equations have to be solved. The detailed theoretical and experimental study on spatial distributions of barium metastable atoms created under the described experimental conditions can be found in [3].

The diffusion coefficients of the barium metastable atoms have been reported in [10]. The value D for the diffusion of Ba triplet metastable atoms in argon at our experimental temperature ($T = 730$ K) is $D = 0.65/p_{\mathrm{Ar}}$ [$\mathrm{cm}^2\ \mathrm{s}^{-1}$], where p_{Ar} (in bar) denotes the argon pressure. Since geometrical cross sections of singlet and triplet metastable barium atoms should be of the same size, one can expect that their diffusion coefficients are not very different. At the experimental temperature, $v^{\mathrm{rel}}_{\mathrm{Ba-Ar}} = 7\times10^4\ \mathrm{cm\ s}^{-1}$ and one can estimate that, for the cylindrical excitation volume with $r_0 = 3$ mm and for pressures above 1 mbar, the diffusion removal rate of the singlet metastable atoms W^{s}_D is small compared with their total collisional depopulation rate $R_{\mathrm{st}} + R_{\mathrm{sp}}$. In the case of the triplet metastables, the collisional depopulation rate R_{ts} is considerably smaller than the collisional depopulation rate for singlets, and the diffusion of ${}^3\mathrm{D}_J$ metastables is expected to be significant, at least up to argon pressures of about 100 mbar.

The back-transfer rates R_{sp} and R_{ts} can be calculated using the data for the corresponding cross sections given in Table 1 and taking into account the principle of detailed balancing. This principle requires the rates R_{ij} and R_{ji} to be in the ratio $R_{ij}/R_{ji} = (g_j/g_i)\exp(-\Delta E_{ji}/kT)$. Accordingly, for the temperature $T = 730$ K the ratios $R_{\mathrm{sp}}/R_{\mathrm{ps}}$ and $R_{\mathrm{ts}}/R_{\mathrm{st}}$ should equal 0.052 and 0.0063, respectively.

The noble gas number density is several orders of magnitude higher than the barium number density. Therefore, the mixing processes induced by collisions with barium ground state atoms are overwhelmed by those which are due to collisions with noble gas atoms. Considering the population of the barium ${}^1\mathrm{P}^0_1$ resonant state within this model, only the energy pooling process (1) is important. An appropriate rate equation for this process can be written as:

$$\frac{\mathrm{d}}{\mathrm{d}t}N_{\mathrm{r}} = 0 = -(\frac{1}{\tau_{\mathrm{r}}} + R)N_{\mathrm{r}} + \frac{k}{2}N_{\mathrm{t}}^2. \qquad (5)$$

Here, k is the energy pooling rate coefficient, τ_{r} is the radiative lifetime of the resonance state and R is the collisional depopulation rate. Factor 1/2 in equation (5) prevents double counting each colliding pair of identical atoms for the case of closed cell energy pooling measurements. This important correction was introduced by Bezuglov *et al.* [11].

The resonant state ${}^1\mathrm{P}^0_1$ decays radiatively to the ground state and to the metastable singlet and triplet states, with the branching ratio $A_{\mathrm{rg}} : A_{\mathrm{rs}} : A_{\mathrm{rt}} = 0.9966 : 0.0025 : 0.0009$ [8]. As reported in [12], the collisional deactivation of ${}^1\mathrm{P}^0_1$ state by noble gas perturbers results in the exclusive production of the ${}^3\mathrm{P}^0_2$ substate [12]. The corresponding cross section for collisions with argon is $2.5 \times 10^{-16}\ \mathrm{cm}^2$. Under our experimental conditions ($N_{\mathrm{Ar}} \approx 3\times10^{17}\ \mathrm{cm}^{-3}$), this yields $R = 5.3 \times 10^6\ \mathrm{s}^{-1}$, which is negligible in comparison with the spontaneous emission rate A_{rg}. Since $1/\tau_{\mathrm{r}} = \sum_i A_{\mathrm{r}i} \cong A_{\mathrm{rg}}$, the rate equation (5) can be rewritten in the following form:

$$\frac{\mathrm{d}}{\mathrm{d}t}N_{\mathrm{r}} = 0 = -A_{\mathrm{rg}}N_{\mathrm{r}} + \frac{k}{2}N_{\mathrm{t}}^2. \qquad (6)$$

Under present experimental conditions the above equation represents a good approximation of the rate equation (5) for cases when A_{rg} is not significantly lowered by the trapping of the resonance radiation.

3.2 Method

For determination of k according to equation (6) the knowledge of N_{t} and N_{r} is needed. In contrast to N_{t}, we were not able to measure N_{r} directly. By comparison of the fluorescence intensities of the Ba resonance and the intercombination line one can obtain the ratio $N_{\mathrm{r}}/N_{\mathrm{p}}$. Unfortunately, we could not measure directly N_{p} either. Nevertheless, we circumvented this problem, and determined N_{p} in an indirect way presented below. In addition to the ratio $N_{\mathrm{r}}/N_{\mathrm{p}}$ we measured the ratio $N_{\mathrm{s}}/N_{\mathrm{t}}$ and the quantity $N_{\mathrm{s}}/N_{\mathrm{p}} + N_{\mathrm{t}}/N_{\mathrm{p}}$. Results of these measurements and the rate equations (2-4) based on the model described in the previous section, provide sufficient data set for the determination of the rate coefficient k.

We assume that the observed fluorescence emerges from an optically thin layer. For our geometry, the spatial distributions of the excited barium atoms, governed by the shape of the laser beam, are axially symmetric. Thus, the population density of the atoms being in a state i can be described as $N_i = N_i^{\mathrm{C}}(z)f_i(r)$. Here, r is a distance from the beam axis in a $x{-}y$ plane, $N_i^{\mathrm{C}}(z)$ is the population density at the axis of the laser beam and $f_i(r)$ is a distribution function normalised to unity at the beam axis ($r = 0$). We assume the number densities to be constant along the beam axis. Furthermore, we assume that the width Δy of the observed fluorescence volume (centrally positioned slab parallel to the beam axis) is small compared with the widths of the population distributions. This means that the population distributions can be taken constant in y direction (within Δy) as well. In this case, only the inhomogeneity in direction of observation is relevant and the emerging fluorescence intensities can be written as:

$$I_{ik} \propto h\,\nu_{ik}L_z\Delta y A_{ik}N_i^{\mathrm{C}}\int f_i(x)\mathrm{d}x. \qquad (7)$$

With known distributions of the excited atoms, the ratio of the fluorescence intensities I_{553}/I_{791} yields the relationship between $N_{\mathrm{r}}^{\mathrm{C}}$ and $N_{\mathrm{p}}^{\mathrm{C}}$ in the centre of the excitation zone:

$$N_{\mathrm{r}}^{\mathrm{C}} = N_{\mathrm{p}}^{\mathrm{C}}\frac{I_{553}}{I_{791}}\frac{\nu_{791}}{\nu_{553}}\frac{A_{\mathrm{pg}}}{A_{\mathrm{rg}}}\frac{\int f_{\mathrm{p}}(x)\mathrm{d}x}{\int f_{\mathrm{r}}(x)\mathrm{d}x}\,. \qquad (8)$$

Applying the rate equation (6) to the number densities obeying our geometry, we obtain the following relation for the rate coefficient k:

$$k = 2A_{\mathrm{rg}}\frac{N_{\mathrm{r}}^{\mathrm{C}}}{(N_{\mathrm{t}}^{\mathrm{C}})^2}\frac{f_{\mathrm{r}}(r)}{f_{\mathrm{t}}^2(r)}\,. \qquad (9)$$

The equation (6) is valid for any position, and its application to the centre of the excitation zone requires that $f_\mathrm{r}(r)$ should be equal to $f_\mathrm{t}^2(r)$. Experimental verification of this fact is presented in Sect. 4.1. By combination of equations (8) and (9), one obtains the expression for k in dependence on the ratio $N_\mathrm{p}^\mathrm{C}/(N_\mathrm{t}^\mathrm{C})^2$. Next, we describe the procedure for determination of this ratio.

The measurement of the rate coefficient k requires sufficient number density of the initial state in process (1). As already mentioned and shown in previous papers [2,3], a significant fraction of barium atoms can be transferred to the metastable triplet state by optical pumping of the $^3\mathrm{P}_1^0$ state and subsequent collisions with noble gas atoms. The process depends strongly on noble gas number density. For argon pressures in the range between 2 mbar and 100 mbar, the diffusion of singlet metastable barium atoms is negligible. In this "medium-pressure" regime, the equations (2), (3) and (4), yield the following expressions for barium number densities in the excitation zone:

$$N_\mathrm{t} = \xi_\mathrm{t} N_\mathrm{p} = \frac{R_\mathrm{ps}R_\mathrm{st} + A_\mathrm{pt}(R_\mathrm{st}+R_\mathrm{sp})}{W_D^\mathrm{t}(R_\mathrm{st}+R_\mathrm{sp}) + R_\mathrm{ts}R_\mathrm{sp}} N_\mathrm{p} \quad (10)$$

$$N_\mathrm{s} = \xi_\mathrm{s} N_\mathrm{p} = \frac{R_\mathrm{ps}(W_D^\mathrm{t} + R_\mathrm{ts}) + A_\mathrm{pt}R_\mathrm{ts}}{W_D^\mathrm{t}(R_\mathrm{st}+R_\mathrm{sp}) + R_\mathrm{ts}R_\mathrm{sp}} N_\mathrm{p} \quad (11)$$

$$N_\mathrm{p} = \frac{\dfrac{\rho B_\mathrm{gp}}{A}}{1 + \dfrac{\rho B_\mathrm{gp}}{A}(1+\xi_\mathrm{s}+\xi_\mathrm{t})} N_0 \quad (12)$$

$$N_\mathrm{s} = \frac{1}{1 + \dfrac{\rho B_\mathrm{gp}}{A}(1+\xi_\mathrm{s}+\xi_\mathrm{t})} N_0. \quad (13)$$

Here, the total number density of barium is denoted by $N_0 = N_\mathrm{s} + N_\mathrm{p} + N_\mathrm{s} + N_\mathrm{r}$, while $A = A_\mathrm{pg} + A_\mathrm{pt} + R_\mathrm{ps} - R_\mathrm{sp}\xi_\mathrm{s}$. As mentioned previously, the population densities are axially symmetric, and their ratios ξ_i follow the same position dependence.

It is obvious that in the case of:

$$\frac{\rho B_\mathrm{gp}}{A}(1+\xi_\mathrm{s}+\xi_\mathrm{t}) \gg 1, \quad (14)$$

equations (10) and (12) can be rewritten in the form:

$$N_\mathrm{t} \approx \frac{\xi_\mathrm{t}}{(1+\xi_\mathrm{s}+\xi_\mathrm{t})} N_0 \quad (15)$$

$$N_\mathrm{p} \approx \frac{1}{(1+\xi_\mathrm{s}+\xi_\mathrm{t})} N_0 \,. \quad (16)$$

For application of our method, the condition (14) had to be fulfilled. In order to find out what are the experimental conditions for which the relation (14) holds, we calculated the average values of ξ_s and ξ_t for the whole excitation volume. We used the data given in Table 1. and applied the principle of detailed balancing for determination of the back transfer rates. For instance, at $\bar{T} = 730$ K and $p_\mathrm{Ar} = 100$ mbar the average values $\bar{\xi}_\mathrm{s}$ and $\bar{\xi}_\mathrm{t}$ are 17.5 and 2780, respectively. In this case, condition (14) is valid if $\rho B \geq 5 \times 10^3\ \mathrm{s}^{-1}$. For barium intercombination line such a pump rate can be achieved with a laser beam having the power about 10 mW and the diameter about 1 cm.

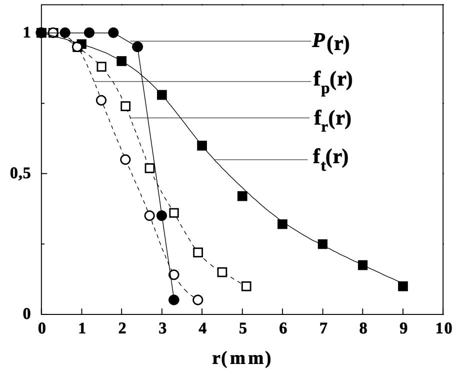

Fig. 3. The spatial distributions of excited Ba atoms; $f_\mathrm{p}(r)$: atoms in the $^3\mathrm{P}_1^0$ state, $f_\mathrm{r}(r)$: atoms in the resonance $^1\mathrm{P}_1^0$ state, $f_\mathrm{t}(r)$: atoms in the triplet metastable state $^3\mathrm{D}_J$. $P(r)$ is the spatial distribution of the pump laser power density.

If the relation (14) holds, and in addition, if $\xi_\mathrm{s} \ll \xi_\mathrm{t}$, than the ground state is strongly depleted and most of the barium atoms are transferred to the triplet metastable state. Under such a conditions, according to equations (15) and (16), the ratio $N_\mathrm{p}/(N_\mathrm{t})^2$ can be replaced with $(1+\xi_\mathrm{s}+\xi_\mathrm{t})/(\xi_\mathrm{t}^2 N_0)$. With this substitution, which comprises the quantities measurable in our experiment, and taking into account the identity $f_\mathrm{r}(r) = f_\mathrm{t}^2(r)$, the combination of equation (8) and equation (9) yields:

$$k = 2\frac{I_{553}}{I_{791}}\frac{\nu_{791}}{\nu_{553}} A_\mathrm{pg} \frac{1+\xi_\mathrm{s}^\mathrm{C}+\xi_\mathrm{t}^\mathrm{C}}{(\xi_\mathrm{t}^\mathrm{C})^2}\frac{1}{N_0}\frac{\int f_\mathrm{p}(x)\mathrm{d}x}{\int f_\mathrm{r}(x)\mathrm{d}x} \,. \quad (17)$$

The above relation shows that for obtaining the rate coefficient k the determination of the spatial distributions, the quantities $\xi_\mathrm{s}^\mathrm{C}/\xi_\mathrm{t}^\mathrm{C}$, $\xi_\mathrm{s}^\mathrm{C}+\xi_\mathrm{t}^\mathrm{C}$, intensity ratio I_{553}/I_{791} and total ^{138}Ba number density N_0, are required. The performed measurements were related to the centre of the excitation zone, and procedures for determination of the required quantities are presented in the following section.

4 Measurements and results

4.1 Spatial distributions of excited barium atoms

Figure 3 shows the spatial distributions. The laser power distribution was measured directly by use of a pin-hole and a photodiode. The distribution of the triplet metastable atoms was measured by shifting the probe beam up to 9 mm from the centre of the pump laser beam. In order to measure the N_p and N_r distributions, the observed layer within the fluorescence zone was discriminated by moving the imaging lens in the y direction and the corresponding intensities of the 791.3 nm and 553.6 nm lines were measured. Applying the Abel inversion method, the respective spatial distribution functions were obtained. Since the pump beam power distribution was homogeneous within the cylinder of radius r_0, one can take that

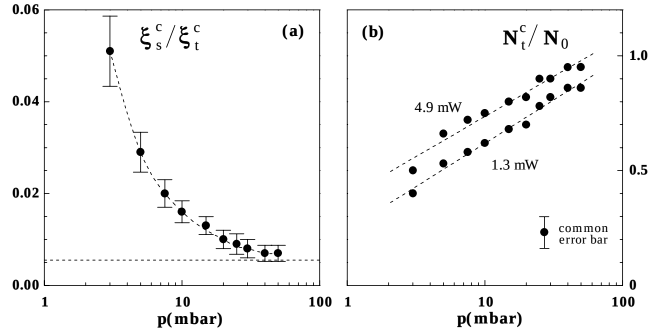

Fig. 4. (a) Singlet to triplet metastable number densities $\xi_{\rm s}^{\rm C}/\xi_{\rm t}^{\rm C} = N_{\rm s}^{\rm C}/N_{\rm t}^{\rm C}$ measured in the excitation zone centre, as a function of argon pressure. The data represent the results obtained for the pump powers of 4.9 and 1.3 mW. Common symbols are used since the difference between the data measured for different laser powers at the same pressure is small compared to the error bar. (b) The ratio of the triplet metastable number density $N_{\rm t}^{\rm C}$ and the total ^{138}Ba number density N_0 as a function of argon pressure at two different pump powers. The data are related to the centre of the excitation zone. The size of the error bars is determined by uncertainties in laser absorption measurements.

$f(y) = f(x) = f(r)$. Note that $f_{\rm r}(r) \approx f_{\rm t}^2(r)$, which confirms the predicted energy pooling rate equation (6).

It should be emphasised that the absorption of the laser radiation was weak (a few percent) as it passed through the barium vapour column (length: $\sim$ 4 cm). Consequently, the number densities of the excited barium atoms are expected to be almost constant along the fluorescence zone. In addition, the measured density distributions show that the number densities of the excited barium atoms are practically constant in y-direction within the observed fluorescence slab (thickness $\Delta y \approx 1$ mm) as well. The geometry realised fairly matches the one predicted in Section 3.2. It simplifies the evaluation of the results, since the volume effects reduce to one-dimensional integrals along the direction of observation.

4.2 Determination of $\xi_{\rm s}/\xi_{\rm t}$ in the excitation zone centre

In our previous work [3] we measured the ratio $\xi_{\rm s}^{\rm C}/\xi_{\rm t}^{\rm C} = N_{\rm s}^{\rm C}/N_{\rm t}^{\rm C}$ in the centre of the excitation zone for argon pressures below 5 mbar. In the same manner, using the laser absorption and laser fluorescence method, we have measured this ratio for argon pressures in the range between 3 mbar and 100 mbar. The diode laser was adjusted to the centre of the Ba 791.3 nm line. In order to determine the $N_{\rm s}^{\rm C}$ and $N_{\rm t}^{\rm C}$ number densities, the respective optical depths of the Ba 582.8 nm and Ba 602.1 nm lines were measured in the centre of the excitation zone by the probe laser beam. The measurements were performed for laser pump powers of 4.9 mW and 1.3 mW. The results are shown in Figure 4a. It was found that the measured ratio $N_{\rm s}^{\rm C}/N_{\rm t}^{\rm C}$ does not depend on the laser pump power applied, which is in accordance with their relationship as predicted by equations (10) and (11). In addition, we measured the $N_{\rm t}^{\rm C}/N_0$ ratio. The barium atom number density N_0 was determined from the absorption of the 791.3 nm line. The diode laser power was reduced down to 3 μW in order to avoid the depletion of the ground state. In this case, the measured ground state number density $N_{\rm g}$ is practically equal to the total number density N_0. The results are given in Figure 4b. In contrast to $N_{\rm s}^{\rm C}/N_{\rm t}^{\rm C}$, the ratio $N_{\rm t}^{\rm C}/N_0$ does show clear dependence on the pump power.

4.3 Determination of $\xi_{\rm s} + \xi_{\rm t}$ in the excitation zone centre

For the determination of $\xi_{\rm s}^{\rm C} + \xi_{\rm t}^{\rm C} = (N_{\rm s}^{\rm C}/N_{\rm p}^{\rm C}) + (N_{\rm t}^{\rm C}/N_{\rm p}^{\rm C})$ we compared the fluorescence intensities I'_{791} and I''_{791} of the optically thin intercombination line for two different laser pump rates $\rho' B_{\rm gp}$ and $\rho'' B_{\rm gp}$. In both cases the argon pressure was varied from 3 mbar to 100 mbar. Since the $I'_{791}/I''_{791} = (N_{\rm p}^{\rm C})'/(N_{\rm p}^{\rm C})''$, equation (12), when written for two different laser powers, provides a set of equations which yields the following expression for the quantity $\xi_{\rm s}^{\rm C} + \xi_{\rm t}^{\rm C}$:

$$\xi_{\rm s}^{\rm C} + \xi_{\rm t}^{\rm C} = \frac{(A_{\rm pg} + A_{\rm pt} + R_{\rm ps})(F-1) + \rho'' B_{\rm gp}(\frac{I'_{791}}{I''_{791}} - 1)}{S R_{\rm sp}(F-1) - \rho'' B_{\rm gp}(\frac{I'_{791}}{I''_{791}} - 1)}, \tag{18}$$

where $S = (\xi_{\rm s}^{\rm C}/\xi_{\rm t}^{\rm C})/(1 + \xi_{\rm s}^{\rm C}/\xi_{\rm t}^{\rm C})$ and $F = I'_{791}\rho''/I''_{791}\rho'$.

The line shapes for our experimental conditions are generally of the Voigt type and the pump rate for the laser beam with the power P and cross section q can be calculated using the following expression, which is valid for the line centre:

$$\rho B_{\rm gp} = \frac{1}{8\pi^{3/2}} \frac{\lambda_{\rm gp}^3 A_{\rm gp}}{{\rm hc}\Delta\nu_D} \frac{g_{\rm p}}{g_{\rm g}} \frac{P}{q} H(a,\,0), \tag{19}$$

Here $H(a,\,0)$ is the Voigt function for the frequency detuning $\Delta\nu = 0$. The parameter a is defined as the ratio of the Lorentzian half-width and the Doppler constant $\Delta\nu_{\rm D}$. For the determination of $H(a,\,\nu)$ we have used the pressure broadening rate for the barium intercombination line broadened by Ar given in [2].

The fluorescence intensity of the intercombination line was measured for laser powers of 1.3 mW and 4.9 mW. The relative intensities I'_{791} and I''_{791} are displayed in Figure 5a. Using these data and the values for $\xi_{\rm s}^{\rm C}/\xi_{\rm t}^{\rm C}$ given in Figure 3a., the $\xi_{\rm s}^{\rm C} + \xi_{\rm t}^{\rm C}$ data in dependence on argon pressure were calculated with the help of equations (18) and (19). The results are shown in Figure 5b.

Apparently, the complicated form of the expression (18) implies a large overall uncertainty of the obtained results for $\xi_{\rm s}^{\rm C} + \xi_{\rm t}^{\rm C}$. For example, the cross section $\sigma_{\rm sp}$ is accurate to about 30%. Nevertheless, for the typical conditions in the presented experiment, equation (18) can be approximately written in a simpler form which shows that the overall uncertainty of the evaluated values for

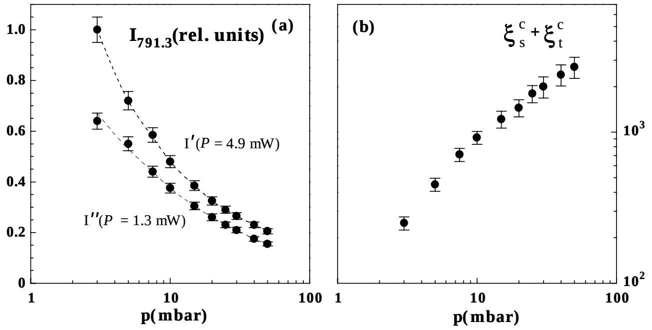

Fig. 5. (a) Relative fluorescence intensities of the optically thin barium intercombination line in dependence on argon pressure and applied laser pump power. (b) Calculated values $\xi_{\rm s}^{\rm C}+\xi_{\rm t}^{\rm C} = (N_{\rm s}^{\rm C}/N_{\rm p}^{\rm C}) + (N_{\rm t}^{\rm C}/N_{\rm p}^{\rm C})$ in dependence on argon pressure.

$\xi_{\rm s}^{\rm C}+\xi_{\rm t}^{\rm C}$ is much lower than one can generally expect. At $p_{\rm Ar} = 30$ mbar, $R_{\rm ps} \approx 10^7\ {\rm s}^{-1}$, *i.e.*, $R_{\rm ps} \gg A_{\rm pg} + A_{\rm pt}$. For pump rates applied ($P = 4.9$ mW; $\rho'' B_{\rm gp} = 2 \times 10^4\ {\rm s}^{-1}$) the second term in the numerator of equation (18) is negligible in comparison with $R_{\rm ps}$, and the numerator practically acquires the form $R_{\rm ps}(F-1)$. Furthermore, at 30 mbar argon pressure the ratio $\xi_{\rm s}^{\rm C}/\xi_{\rm t}^{\rm C} \ll 1$, which means that the quantity S can be replaced with $\xi_{\rm s}^{\rm C}/\xi_{\rm t}^{\rm C}$. Therefore, under typical experimental conditions a good approximative form of equation (18) can be written as follows:

$$\xi_{\rm s}^{\rm C}+\xi_{\rm t}^{\rm C} \approx \frac{F-1}{\dfrac{\xi_{\rm s}^{\rm C}}{\xi_{\rm t}^{\rm C}}\dfrac{R_{\rm sp}}{R_{\rm ps}}(F-1) - \dfrac{\rho'' B_{\rm gp}(I'_{791}/I''_{791}-1)}{R_{\rm ps}}}. \quad (20)$$

Since the ratio $R_{\rm ps}/R_{\rm sp}$ is determined by the principle of the detailed balance, the error due to uncertainty of $R_{\rm ps}$ propagates *via* the second term in the denominator only. This term is about 2 times smaller than the first term, and the inaccuracy in $R_{\rm ps}$ contributes to the overall uncertainty of $\xi_{\rm s}^{\rm C}+\xi_{\rm t}^{\rm C}$ with about 15%. Uncertainty of the ratio $R_{\rm ps}/R_{\rm sp}$ for the experimental temperature range is about 5%. Taking into account the statistical accuracy of the $\xi_{\rm s}^{\rm C}+\xi_{\rm t}^{\rm C}$ which is 10%, the overall uncertainty of this quantity is estimated to 30%.

4.4 Experimental conditions

The rate coefficient k was measured at $p_{\rm Ar} = 30$ mbar. At this pressure and for the applied pump powers, the requirement given by (14) is fulfilled. Indeed, using (19) and the data displayed in Figure 4b, one can check that for the pump powers 4.9 mW and 1.3 mW the product $(\rho B_{\rm gp}/A) \times (1 + \xi_{\rm s}^{\rm C} + \xi_{\rm t}^{\rm C})$ is about 40 and 11, respectively. Consequently, the $N_{\rm p}/N_{\rm t}^2$ ratio in equation (9) can be replaced with $(1+\xi_{\rm s}^{\rm C}+\xi_{\rm t}^{\rm C})/(\xi_{\rm t}^{\rm C})^2 N_0$, *i.e.*, equation (17) is applicable for the evaluation of the rate coefficient. The error in determination of the rate coefficient k, introduced by this replacement amounts to 5%. The pressure $p_{\rm Ar} = 30$ mbar was chosen because the intensity of the energy pooling resonant line shows its maximum at this pressure. This behaviour is in accordance with our model. Namely, if the buffer gas pressure increases, the laser pump rate decreases due to pressure broadening of the pump line. As the consequence, the initial energy pooling state, *i.e.* the $N_{\rm t}$ state, is populated with smaller efficiency.

The ^{138}Ba atom number density N_0 was varied between $2 \times 10^{10}\ {\rm cm}^{-3}$ and $6 \times 10^{10}\ {\rm cm}^{-3}$. The lower density was determined by the sensitivity of our apparatus, while the higher value was limited by the required optical thickness of the resonance line. From an extrapolation of the data for the barium number density in dependence on temperature, which are given in reference [3], the temperature was estimated to be in the range between 700 and 760 K.

4.5 Measurements of the I_{553}/I_{791} ratio

The intercombination line was optically thin in the whole vapour column. Therefore, the measured intensity is proportional to the intercombination line intensity emerging from the fluorescence zone:

$$I_{791}^{\rm meas} \propto I_{791} \propto h\,\nu_{791}\Delta y\, L_z\, A_{\rm pg}\, N_{\rm p}^{\rm C} \int_{-R}^{d} f_{\rm p}(x){\rm d}x. \quad (21a)$$

On the other hand, the resonant radiation emerging from the fluorescence zone, characterised by strong depletion of the ground state, had to pass an optically thick layer of ground state atoms outside the fluorescence volume. Thus the measured resonant fluorescence intensity reads:

$$I_{553}^{\rm meas} \propto h\,\nu_{553}\Delta y\, L_z\, A_{\rm rg} N_{\rm r}^{\rm C} \int_{-R}^{d} f_{\rm r}(x){\rm e}^{-\sigma\int_x^d N_{\rm g}(x'){\rm d}x'}{\rm d}x. \quad (21b)$$

where σ is the effective cross section for the photoabsorption. The exponential term in the above expression represents the escape probability for the resonance radiation. From Figure 3 one can see that more than 80% of the barium atoms in the resonance state are confined in the cylinder with a radius $r_0 = 3$ mm. The number density of the ground state atoms is reversed to the triplet metastable number density ($N_{\rm g}(x) = N_{\rm g}^{\rm C}[1-f_{\rm t}(x)]$) and, according to data given in Figure 3, the average ground state number density $N_{\rm g}$ in that volume is about 10% of the total barium number density ($\overline{N}_{\rm g} \approx 4 \times 10^9\ {\rm cm}^{-3}$). The corresponding peak absorption coefficient k_0 of the resonance line amounts $\sim 0.1\ {\rm cm}^{-1}$, which means that the considered volume can be fairly regarded as optically thin. The remaining 20% of the atoms in the resonance state spread to the region where the ground state number density begins to rise towards its total value. Nevertheless, the density of the resonance atoms is practically exhausted by the point where the ground state number density reaches about half of its total value ($N_{\rm g} \approx N_0/2 = 2 \times 10^{10}\ {\rm cm}^{-3}$). The average value for the peak absorption coefficient in this region amounts to about $0.5\ {\rm cm}^{-1}$. The latter still does not cause serious trapping of resonance radiation. The resonance fluorescence is practically restricted to the optically thin cylinder with radius $r_1 \approx 0.5$ cm. Thus the upper and lower limits in the x integral in expression (21b) can be replaced with r_1 and $-r_1$, respectively. Furthermore, for the fluorescence that is confined to a small region at the centre of the heat pipe ($r_1 \ll d$), the lower limit in the x' integral can be replaced by zero and equation (21b) reads:

$$I_{553}^{\rm meas} \propto \alpha I_{553} \propto h\,\nu_{553}\Delta y\, L_z A_{\rm rg} N_{\rm r}^{\rm C}\alpha \int_{-r_1}^{r_1} f_{\rm r}(x){\rm d}x, \tag{22}$$

where the factor α, given by:

$$\alpha = {\rm e}^{-\sigma\int_0^d N_{\rm g}(x'){\rm d}x'}, \tag{23}$$

represents an absorption correction for the resonance radiation that emerges from the optically thin source and is absorbed along the optically thick path between the source and the observer.

We have replaced the x' integral in the above equation by a product $N_{\rm g}\bar{d} = N_0\bar{d}$, where $\bar{d}$ labels the mean optical path from the fluorescence zone to the end of the vapour column in the observation direction. The uncertainty of the mean optical path was estimated to be ~ 0.5 cm, which includes the inhomogeneous ends of the absorbing layer and slight differences between different optical paths within the small observation angle (0.05 rad) around the x-axis. The absorption factor α for the resonance line intensity was calculated according to the following relation:

$$\alpha = \frac{\int H(a,\,\nu)\exp[-k_0\bar{d}\,H(a,\,\nu)]{\rm d}\nu}{\int H(a,\,\nu){\rm d}\nu} \tag{24}$$

where the peak-absorption coefficient

$$k_0 = (N_{\rm g} g_{\rm g}\lambda_{\rm gr}^3 A_{\rm rg}\sqrt{M_{\rm Ba}/2kT})/(8\pi^{3/2}g_{\rm r}).$$

Since only ^{138}Ba atoms were excited, the absorption factor was calculated as self-absorption of the single line. The ^{138}Ba resonance radiation emitted in the line wings can be absorbed by hyperfine components of other Ba isotopes. For our experimental conditions though, taking into account the isotope structure of the Ba resonance line would affect α by less than 0.25%. In order to minimize the uncertainty related to the inhomogeneous absorption layer, we measured the intensity ratio $\eta = I_{553}^{\rm meas}/I_{791}^{\rm meas}$ for two alignments of the pump laser beam. In the first case, the beam was shone coaxially through the long arm of the stainless-steel cell, while in the second case, the pump beam was shifted by 1 cm in the direction of observation parallel to the beam axis. We estimated the mean lengths of the absorbing barium ground-state layers in the first and the second cases to be 2 cm and 1 cm, respectively. The data for the measured intensity ratio η and the corrected values $\eta^{\rm corr} = \eta/\alpha$, are displayed in Figure 6. The values for $\eta^{\rm corr}$ correspond to the ratio I_{553}/I_{791} for the radiation from the optically thin observed slab. Although the uncertainty in the optical path d itself is relatively large, the difference between the paths in these two measurements is exactly known. The mutual agreement of the corrected η values for two cases at hand, shows that the mean values of d are well estimated. These estimates were obtained by an iterative procedure, where two conditions were required: exact difference between the mean optical paths should be 1 cm, and, the corresponding corrections for absorption should be in agreement. In this way, the uncertainties due to absorption of the resonance radiation at the path d can be eliminated in evaluation of the resonance to intercombination line intensity ratio.

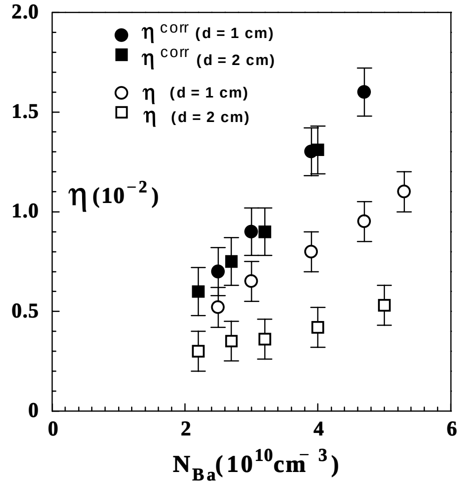

Fig. 6. The measured $\eta = I_{553}^{\rm meas}/I_{791}^{\rm meas}$ and the absorption corrected $\eta^{\rm corr} = \eta/\alpha$ fluorescence intensity ratios against ^{138}Ba number density.

4.6 Result for the rate coefficient k

The rate coefficient k was determined by use of the relation (17). The value for $(1 + \xi_{\rm s}^{\rm C} + \xi_{\rm t}^{\rm C}/(\xi_{\rm t}^{\rm C})^2$ was found to be $1/1900 \pm 10\%$ and the ratio of the averaged spatial distributions was 0.8. With the data for $\eta^{\rm corr}$ displayed in Figure 6 we obtained the set of the k data corresponding to the temperature range between 700 K and 760 K. The average value for the energy pooling rate coefficient was found to be:

$$k = 1 \times 10^{-10}\ {\rm cm}^3\ {\rm s}^{-1}. \tag{25}$$

The statistical accuracy of this result is about 30%. Including the systematic error in determination of the N_0, and the inaccuracy of the data used for evaluation, we estimate the overall uncertainty to be about 70%. The corresponding cross section $\sigma = k/v_{\rm Ba-Ba}^{\rm rel}$ is 3.2×10^{-15} cm^2.

5 Discussion and conclusion

We have measured the rate coefficient for the energy pooling process Ba (^{3}D$_J$) + Ba (^{3}D$_J$) → Ba(^{1}S$_0$) + Ba(^{1}P$_1^0$). The barium triplet metastable state was populated by radiative and collisionally enhanced relaxation of the laser-excited ^{3}P$_1^0$ state. In this way, most barium ground state atoms in the excitation volume were transferred to the metastable triplet state, and a maximum energy pooling signal, *i.e.* the fluorescence intensity of the barium resonance line, was achieved. The measurements were performed at ^{138}Ba ground state number density $N_0 \approx 4\times10^{10}$ cm^{-3}. Due to strong depletion of the barium ground state in the excitation zone ($N_\mathrm{g}^\mathrm{C} \approx 4 \times 10^9$ cm^{-3}) the trapping of resonance radiation was avoided. The typical values for the measured ratios $N_\mathrm{p}^\mathrm{C}/N_\mathrm{t}^\mathrm{C}$, and $N_\mathrm{r}^\mathrm{C}/N_\mathrm{p}^\mathrm{C}$ were 5×10^{-4} and 10^{-5}, respectively. Since $N_\mathrm{t}^\mathrm{C} \approx 4\times10^{10}$ cm^{-3}, the typical value for the population density of the resonance state N_r^C was 2×10^2 cm^{-3}.

Taking the fine structure of the $^3\mathrm{D}_J$ state into account, the measured rate coefficient k is the sum of six particular contributions. These contributions are due to collisions between barium triplet metastables in different sublevels. In the present experiment, the triplet metastable state was completely mixed and we were not able to distinguish the different rate coefficients. However, the maximum values for each particular contribution can be derived. The measured rate coefficient is an average value which comprises the particular contributions due to $^3\mathrm{D}_J + {}^3\mathrm{D}_{J'}$ collisions. Following the basic energy pooling equation for the considered system, the relation between the measured overall coefficient k and six particular contributions can be written as:

$$\frac{k}{2}N^2(^3\mathrm{D}_J) = \frac{k_{11}}{2}N^2(^3\mathrm{D}_1) + k_{12}N(^3\mathrm{D}_1)N(^3\mathrm{D}_2) + \ldots\ldots + \frac{k_{33}}{2}N^2(^3\mathrm{D}_3). \tag{26}$$

With equilibrium population distributions of the $^3\mathrm{D}_J$ sublevels at the experimental temperature, taken into account, equation (26) reads:

$$\frac{k}{2} = \frac{k_{11}}{2}0.12 + k_{12}0.14 + \ldots\ldots + \frac{k_{33}}{2}0.07. \tag{27}$$

As mentioned in the Introduction, the most interesting case is the quasi-resonant case of the $^3\mathrm{D}_1 + {}^3\mathrm{D}_1$ collisions. According to our experimental result for k and equation (27), we estimate the maximum value for the rate coefficient k_{11} to be $k/0.12 = 8.3 \times 10^{-10}$ cm^3 s^{-1}, and the corresponding cross section $\sigma_{11} = 2.7 \times 10^{-14}$ cm^2. This value indicates that the investigated energy pooling process could be governed by the long range interaction potentials between the triplet metastable atoms. We believe that this result might be a helpful indication for further theoretical investigations on this subject.

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