Z. Phys. D – Atoms, Molecules and Clusters 22, 449–450 (1992)

Atoms, Molecules
Zeitschrift für Physik D and Clusters

Fine structure excitation transfer between the potassium 4^2P states induced by collisions with caesium atoms

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Received 8 August 1991

Abstract. Applying diode-laser resonant fluorescence method, the cross sections for the excitation energy transfer of the collisional process $K^*(4^2P_{1/2}) + Cs(6^2S_{1/2}) \leftrightarrow K^*(4^2P_{3/2}) + Cs(6^2S_{1/2})$ have been measured. The values we have obtained are $\sigma(1/2 \rightarrow 3/2) = 77$ Å^2 and $\sigma(3/2 \rightarrow 1/2) = 48$ Å^2. These results complete the sequence of data for the fine-structure mixing of the first-resonance states of alkali atoms colliding with the ground-state caesium atoms.

PACS: 34.50; 32.00.

Recently it has been reported on the cross sections for the fine-structure mixing of the $^7Li^*(2^2P)$ states induced by collisions with the ground-state caesium atoms [1]. As the contribution to the data on these processes in mixed alkali systems, here we report the cross sections for analogous processes in K* – Cs collisional system.

High purity caesium metal with a small amount of potassium was contained in the side-arm of a cylindrical cell made of Pyrex glass. Temperature of the cell was kept constant (573 K) during the measurement, while the temperature of the side-arm was varied in the range from 470 K to 550 K. The caesium number density N_{Cs} was controlled spectroscopically by a white-light absorption measurements in the blue wing of the self-broadened resonance Cs $D2$ line at 852.1 nm [2]. The K number density was determined using the curve of growth method for optical thin lines. Since the number density of K was about five orders of magnitude lower than that of Cs, the contributions due to K* – K collisions could be neglected. Therefore, the fine-structure mixing process $K^*(4^2P_{3/2}) \leftrightarrow K^*(4^2P_{1/2})$ was induced only by collisions with Cs atoms.

K atoms in Cs vapours were excited to the $4^2P_{3/2}$ or $4^2P_{1/2}$ state by single mode frequency stabilized diode laser (Hitachi 7802 series, wavelength 785 nm at 25 °C, linewidth about 25 MHz, maximal power 5 mW). The fluorescent zone was imaged onto the entrance slit of the 1 m McPherson monochromator. The direction of the observation was perpendicular to the laser beam. The monochromator slit-widths were 300 μm providing the resolution of about 0.3 nm. The laser was tuned either to the centre of K $D1$ or to the centre of K $D2$ line, while the total intensities of both the sensitized and direct fluorescence were detected using the broad-band adjusted monochromator supplied with the RCA S-20 photomultiplier. Following Krause [3] the sensitized-to-direct fluorescent intensity ratios are:

$$\eta_a = I(D1)/I(D2) \quad \text{and} \quad \eta_b = I(D2)/I(D1). \tag{1}$$

The obtained intensity ratios plotted against Cs number density are shown in Fig. 1. Data analysis was performed applying a simple three-level model which includes $K(^2S_{1/2})$, $K^*(^2P_{1/2})$ and $K^*(^2P_{3/2})$ states. If $K^*(4^2P_{3/2})$ (state 2) is optically excited, then the steady-state equation for the collisionally populated $K^*(4^2P_{1/2})$ state

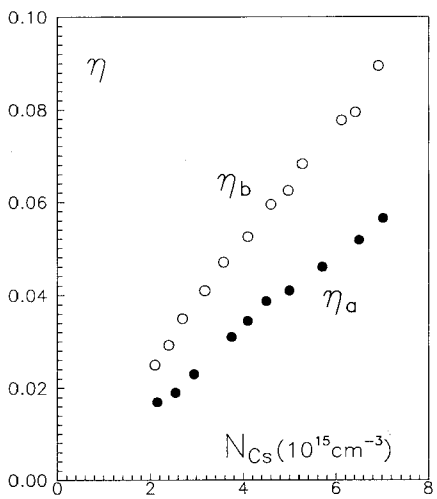

Fig. 1. Sensitized-to-direct fluorescent intensity ratios for the fine-structure mixing of the $K^*(4^2P)$ state due to collisions with Cs ground-state atoms. See text for further explanations

(state 1) is:

$$\dot{N}_1 = 0 = DN_2 - AN_1 - CN_1, \tag{2}$$

which yields the population density ratio

$$N_1/N_2 = D/(A+C). \tag{3}$$

Analogously, for optically populated $K^*(^2P_{1/2})$ one obtains

$$N_2/N_1 = C/(A+D). \tag{4}$$

Here, A denotes the spontaneous emission rate, while C and D are collisional rates for $K^*(4^2P_{1/2}) \rightarrow K^*(4^2P_{3/2})$ and $K^*(4^2P_{3/2}) \rightarrow K^*(4^2P_{1/2})$ transitions, respectively. According to gas kinetics the collisional rate is defined as the product $v\sigma N_p$, where v is average relative velocity of the colliding atoms ($v = \sqrt{8kT/\pi\mu}$, μ is reduced mass), σ denotes the corresponding cross section, and N_p is the number density of the atoms inducing the transition.

Assuming the optically thin conditions and absence of the radiation trapping the fluorescence intensity ratios reduce to population density ratios of the excited levels, i.e.:

$$\eta_a = N_1/N_2 \quad \text{and} \quad \eta_b = N_2/N_1. \tag{5}$$

Combination of (1), (3), (4) and (5), with $\eta_a, \eta_b \ll 1$, which has happened to be the case, yields the expressions for the cross sections

$$\sigma(3/2 \rightarrow 1/2) = A\eta_a/(vN_{Cs}), \tag{6a}$$

$$\sigma(1/2 \rightarrow 3/2) = A\eta_b/(vN_{Cs}) \tag{6b}$$

The measurements have been done in the range of K number densities from $8 \cdot 10^9$ to $3 \cdot 10^{10}$ cm^{-3}. In order to avoid the radiation trapping effects the measurements have to be carried out at K densities lower than some critical value. According to Holstein's theory [4] and for geometry of our experiment, we have estimated this value to be about $5 \cdot 10^{10}$ cm^{-3}. Therefore, the radiation trapping effects in present experiment can be regarded negligible. Using the experimental data (see Fig. 1) and (6) with $A = 3.8 \cdot 10^7$ s^{-1} [5], we have obtained the following values:

$$\sigma(3/2 \rightarrow 1/2) = 48 \text{ Å}^2,$$

$$\sigma(1/2 \rightarrow 3/2) = 77 \text{ Å}^2.$$

Statistical accuracy of these results is about 5%. The total error should include uncertainty due to polarization effects which have not been investigated in this work. As shown by Ballagh and Cooper [6], the collisions destroy the polarization anisotropy and, in the case when collisional broadening parameter is much larger than the natural line width, the polarization anisotropy disappears. Applying the results [6] we have estimated the error due to polarization effects to be at most $\pm 20\%$. In spite of that, the ratio of the experimental cross sections is in satisfactory agreement with the prediction of the detailed balancing, which requires

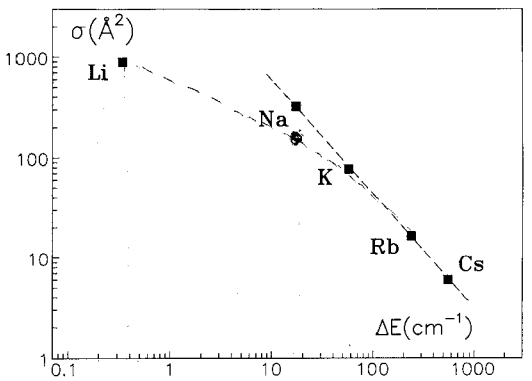

Fig. 2. The relationship between cross sections $\sigma(1/2 \rightarrow 3/2)$ and the fine-structure splitting of the first resonance levels for the alkali atoms colliding with ground-state caesium atoms

$$\sigma(1/2 \rightarrow 3/2)/\sigma(3/2 \rightarrow 1/2) = (g_2/g_1)\exp(-\Delta E/kT). \tag{7}$$

Here, g_2 and g_1 are degeneracies of the $4^2P_{3/2}$ and $4^2P_{1/2}$ potassium states and ΔE (57.7 cm^{-1}) is the corresponding fine-structure splitting. For the temperature of 573 K, (7) yields 1.73 which should be compared with experimental ratio of 1.6.

Figure 2 shows $\sigma(1/2 \rightarrow 3/2)$ plotted against ΔE for all alkali-caesium pairs. The data for Li* – Cs, Na* – Cs, Rb* – Cs and Cs* – Cs are taken from [1, 7–9], respectively. The data were obtained at different temperatures, but comparison of data makes sense because the corresponding temperatures lie in the relatively narrow range (520 ± 50 K). As one can see, all the data with exception of Li* – Cs case, fall on the straight line. The existing theoretical results concerning the excitation energy transfer are scarce and uncertain [9], and we hope that the completeness of the presented sequence of the experimental data would serve as firm basis for further theoretical investigations.

We would like to thank the Ministry of science, technology and informatics (Republic of Croatia) and NIST/NBS (USA) for financial support.

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