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Energy-pooling collisions in Rb-Cs vapor mixture

\[ \text{Rb}(5P_J) + \text{Cs}(6P_{3/2}) \rightarrow \text{Rb}(5S_{1/2}) + \text{Cs}(nl'J') \]

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Rate coefficients for energy-pooling (EP) collisions \( \text{Rb}(5P_J) + \text{Cs}(6P_{3/2}) \rightarrow \text{Rb}(5S_{1/2}) + \text{Cs}(nl'J') \) have been measured. Atoms were excited to \( \text{Rb}(5P_J) \) and \( \text{Cs}(6P_{3/2}) \) states using two single-mode diode lasers. To isolate the heteronuclear contribution in the fluorescence spectrum, a double-modulation technique has been adopted. The excited-atom density and spatial distribution are mapped by monitoring the absorption of a counterpropagating single-mode diode laser beam, tuned to \( \text{Rb}(5P_J \rightarrow 7S_{1/2}) \) and \( \text{Cs}(6P_{3/2} \rightarrow 8S_{1/2}) \) transitions respectively, which could be translated parallelly to the pump beams. The excited atom densities are combined with the measured fluorescence ratios to determine cross sections for the EP processes. It was found that \( \text{Rb}(5P_{1/2}) + \text{Cs}(6P_{3/2}) \) collisions are more efficient than \( \text{Rb}(5P_{3/2}) + \text{Cs}(6P_{3/2}) \) collisions for populating \( \text{Cs}(4F_{7/2}) \), while the opposite is true for populating \( \text{Cs}(4F_{9/2}) \).

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In energy-pooling (EP) collisions\(^{[1-5]}\) two excited atoms produce one ground-state and one very-excited-state atom under the condition that the total internal energy is preserved in the limit of \( kT \). Therefore the energy of the final excited level must be \( E = E_1 + E_2 + \Delta E \), where \( E_1 \) and \( E_2 \) are the energies of the two laser-populated levels and \( \Delta E \approx kT \). This condition imposes severe limitations on the number of possible EP resonances for a given atom. The simultaneous excitation of atoms of two different species in a vapor mixture increases the number of possible “quasi” resonances and it proposes new interesting level scheme combinations. Moreover, it opens the possibility of the theoretical and experimental studies on the interatomic potentials between different atoms in an important range of interatomic distance.

Namiotka et al.\(^{[2]}\) reported experimental rate coefficients for EP process \( K(4P_J + K(4P_J) \rightarrow K(4S) + K(nl)) \) with product states \( nl = 5P, 6S, \) and \( 4D \), and found that the angular momentum propensity rules can play a role in the EP processes.

Here we report the results of an experimental investigation of EP processes

\[ \text{Rb}(5P_J) + \text{Cs}(6P_{3/2}) \rightarrow \text{Rb}(5S) + \text{Cs}(8S, 4F). \]  

The \( \text{Rb}(5P_J) \) fine-structure splitting is large enough to allow a separate study of the \( \text{Rb}(5P_{1/2}) + \text{Cs}(6P_{3/2}) \) and \( \text{Rb}(5P_{3/2}) + \text{Cs}(6P_{3/2}) \) collision processes. Thus the study of angular momentum in the EP processes is independent of energy deficits (see Fig. 1).

It is evident that the double resonant excitation produces a very rich spectra. These spectra are more crowded because of the separate contributions of the homonuclear and of the heteronuclear collisions. In order to measure the heteronuclear rate constants of reaction (1) it is therefore necessary to isolate the signal of reaction (1) from all the others.

We have solved the problem by adopting an intermediate modulation technique that can be used every time heteronuclear processes take place. The two laser beams are modulated at two different frequencies \( \omega_1 \) and \( \omega_2 \), and the fluorescence is phase detected at \( \omega_1 + \omega_2 \). This procedure filters out the homonuclear part of the spectrum, which will have only one of the two modulation frequencies, and it transmits the heteronuclear contribution, which will have components modulated at the sum frequency.

By assuming that other processes, such as multiphoton excitation, atomic ionization etc., are negligible (the experimental conditions are chosen to minimize them). For pumping \( \text{Rb}(5P_J) \) and \( \text{Cs}(6P_{3/2}) \) states, the steady state rate equations for reaction (1) is

\[ [\text{Cs}nlJ'(\tau'_{nl})] = k_{nlJ'}[\text{Rb}5P_J(\tau)][\text{Cs}6P_{3/2}(\tau)] , \]

where \( k_{nlJ'} \) is the EP rate coefficient, \( [\text{Cs}nlJ'(\tau')], [\text{Rb}5P_J(\tau')], \) and \( [\text{Cs}6P_{3/2}(\tau')] \) are the densities in atomic levels \( \text{Cs} (nl'J'), \text{Rb} (5P_J), \) and \( \text{Cs} (6P_{3/2}), \) respectively, \( \tau'_{nlJ'} \) is the effective lifetime for atoms in the states \( \text{Cs}(nlJ') \). However, in the present case, the trapping on all transitions of interest is negligible (i.e., \( \tau'_{nlJ'} = \tau_{nlJ'} \)).

Note that in the absence of quenching collisions and

Fig. 1. (a) Energy levels of rubidium and cesium transitions. Wavelengths are given in nanometer; (b) Cesium atomic levels lying near the \( \text{Rb}(5P_J) + \text{Cs}(6P_{3/2}) \) energies (dashed lines).
radiation trapping, $[\tau_{nl'j'}]^{-1}$ is just the sum of the Einstein A coefficients for transitions connecting state $nlj'$ to all lower levels.

Under the low density of ground-state atoms, a complete model for cesium EP collisions has been presented in Ref. [1], in which the EP rate coefficient in terms of the measured fluorescence ratios is given. EP collisions (reaction (1)) can be treated analogously and so here we only quote the result

$$k_{nlj} = \frac{I_{nlj} - n'l'j'/(\varepsilon_{nlj} - n'l'j')} {I_{6P_{3/2} - 6S_{1/2}}} = \frac{\lambda_{nlj} - n'l'j'} {\lambda_{6P_{3/2} - 6S_{1/2}}} \times \frac{\Gamma_{nlj} - n'l'j'} {\Gamma_{6P_{3/2} - 6S_{1/2}}} \times \frac{1} {\tau_{nlj}} \int_{-R}^{R} \frac{[\text{Cs}_{6P_{3/2}}(x)] [\text{Cs}_{6P_{3/2}}(x)] dx} {T_{6P_{3/2} - 6S_{1/2}}},$$

where $I_{nlj} - n'l'j'$ is the fluorescence intensity corresponding to the transition $nlj' \rightarrow n'l'j'$, $\varepsilon_{nlj} - n'l'j'$ the detection system efficiency (including effects due to the photomultiplier, monochromator grating, and any lens used) at the frequency of interest, and $\lambda_{nlj} - n'l'j'$ the transition wavelength; $R$ is the radius; $\Gamma_{nlj} - n'l'j'$ the average probability that a photon emitted in the detection direction passes through the vapor between its point of origin and the cell walls without being absorbed. It is related to the frequency-dependent absorption cross section (including hyperfine structure), the density, and spatial distribution of atoms in the lower level transition, and is given explicitly in Eqs. (8) — (11) of Ref. [1]. We note that in the experiment, $T$ is approximately equal to 1 except for the $6P_{2/3} \rightarrow 6S_{1/2}$ resonance transition. The resonance broadening rates of Ref. [6] were used to calculate the $T_{6P_{3/2} - 6S_{1/2}}$. Experimentally, the measured quantities are $I_{nlj} - n'l'j'$, $\varepsilon_{nlj} - n'l'j'$, $\lambda_{6P_{3/2} - 6S_{1/2}}$, $\varepsilon_{6P_{3/2} - 6S_{1/2}}$, $[\text{Rb}_{6P_{3/2}}(x)]$, and $[\text{Cs}_{6P_{3/2}}(x)]$.

Figure 2 shows the set-up of the experiment. A Rb-Cs alloy was confined in a cylindrical cell with the inner diameter of 1.2 cm and the length of 6 cm. The cell was sealed after baking and evacuating until a vacuum of $10^{-4}$ Pa was reached. The alloy in the cell consisted of 70% rubidium and 30% cesium, and provided approximately equal atom densities according to Raoult’s law. The cell was placed inside an oven. The temperatures of the cell ranging from 337 to 347 K were measured with thermocouples. The total density in the cell is of the order of $10^{14}$ cm$^{-3}$. This procedure gives reasonably comparable vapor pressures and avoids the problems connected to secondary processes induced by too high vapor pressure of one of the two elements.

Two lasers (Toptica DL100, linewidth of $\sim 1$ MHz) were used to excite rubidium (laser 1) and cesium (laser 2) respectively. Laser 1 was a single mode diode laser with a maximum output power of 40 mW, laser 2 is also a single mode diode laser with 70 mW. The wavelengths were 794.8 (or 780.0) and 852.3 nm. A chopper modulated the two beams at the frequencies $\omega_1$ and $\omega_2$. The lock-in amplifier (Stanford research system SR830) was tuned to the sum frequency. The resonance fluorescence signals were observed with an infrared sensitive charge coupled device (CCD) camera during the alignment of the laser beams along the cell axis. We tuned the laser frequencies to maximize Rb($5P_1 \rightarrow 5S_{1/2}$) and Cs($6P_{3/2} \rightarrow 6S_{1/2}$) fluorescence from the observation region. At the lower densities, the attenuation of the laser beams along the cell length could generally be neglected even when the laser was tuned directly to resonance.

Fig. 2. (a) Experimental setup; (b) Inset showing the cell geometry and the region (shaded) from which fluorescence was detected.

A single-mode diode laser was used to probe the density and spatial distributions of the atoms in Rb($5P_{1/2}$) level. The power of the probe laser was cut down to $\sim 1$ μW with neutral density filters. We directly measure the $5P_{1/2}$ density by scanning the probe laser over the Rb($5P_1 \rightarrow 7S_{1/2}$) transition and monitoring its transmission using another photomultiplier tube (PMT-2). To measure the spatial dependence $[\text{Rb}_{5P_{3/2}}(x)]$ across the diameter of the cell, the probe laser was stepped across the cell parallel to the pump beam using the translating mirror as shown in Fig. 2.

The transmitted intensity of the probe laser beam through a length $L$ of the vapor is given by

$$I_\nu = I_\nu(0) e^{-k_{7S_{1/2} \rightarrow 5P_{1/2}}(\nu)L}$$

for the light of frequency $\nu$. Here $I_\nu(0)$ is the incident intensity and $k_{7S_{1/2} \rightarrow 5P_{1/2}}(\nu)$ is the frequency-dependent
Table 1. Rate Coefficients for the EP Collisions Rb(5P_J)+Cs(6P_3/2) → Rb(5S)+Cs(nl_J')

<table>
<thead>
<tr>
<th>nl_J'</th>
<th>T (°C)</th>
<th>n_o S_{1/2} (10^{11} \text{ cm}^{-3})</th>
<th>Ratio 1</th>
<th>Monitored Trans. nl_J' → n'l_J'</th>
<th>Ratio 2</th>
<th>\gamma_{nl_J' → n'l_J'}</th>
<th>\Gamma_6^{e}P_{3/2} → \delta S_{1/2} (s^{-1})</th>
<th>\kappa_{nl_J'} (\text{cm}^{3}\text{s}^{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>4F_{5/2}</td>
<td>64</td>
<td>2.72</td>
<td>(1.41 \times 10^{3})^{-1}</td>
<td>4F_{5/2} → 5D_{3/2}</td>
<td>6.10 \times 10^{-8}</td>
<td>0.877</td>
<td>4.53 \times 10^{0}</td>
<td>2.64 \times 10^{-10}</td>
</tr>
<tr>
<td>74</td>
<td>(3.79 \times 10^{3})^{-1}</td>
<td>3.92 \times 10^{-7}</td>
<td>1.94 \times 10^{4}</td>
<td>2.71 \times 10^{-10}</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>4F_{7/2}</td>
<td>64</td>
<td>2.72</td>
<td>(1.41 \times 10^{3})^{-1}</td>
<td>4F_{7/2} → 5D_{5/2}</td>
<td>3.30 \times 10^{-8}</td>
<td>0.936</td>
<td>4.53 \times 10^{4}</td>
<td>1.35 \times 10^{-10}</td>
</tr>
<tr>
<td>74</td>
<td>(3.79 \times 10^{3})^{-1}</td>
<td>2.16 \times 10^{-7}</td>
<td>1.94 \times 10^{4}</td>
<td>1.41 \times 10^{-10}</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>8S_{1/2}</td>
<td>64</td>
<td>2.72</td>
<td>(1.41 \times 10^{3})^{-1}</td>
<td>8S_{1/2} → 6P_{3/2}</td>
<td>1.93 \times 10^{-7}</td>
<td>0.380</td>
<td>4.53 \times 10^{4}</td>
<td>15.2 \times 10^{-10}</td>
</tr>
<tr>
<td>74</td>
<td>(3.79 \times 10^{3})^{-1}</td>
<td>1.08 \times 10^{-6}</td>
<td>1.94 \times 10^{4}</td>
<td>13.5 \times 10^{-10}</td>
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<td></td>
</tr>
</tbody>
</table>

An estimate of the statistical errors in the measured rate coefficients can be obtained by Eq. (3). Fluorescence ratios probably have an uncertainty as much as 25% because the nl_J' → n'l_J' fluorescence signals were weak. We estimate the uncertainty in the ratio \( \int_{-R}^{R} [\text{Cs}_{6P_{3/2}}(x)]dx / \int_{-R}^{R} [\text{Rb}_{5P_{J}}(x)] \int_{-R}^{R} [\text{Cs}_{6P_{3/2}}(x)]dx \) to be approximately 15%. Values of \( \gamma_{nl_J' → n'l_J'} \) is probably good to within 10%, while the values of \( \Gamma_6^{e}P_{3/2} → \delta S_{1/2} \) are accurate to at least 10% because the trapping is much less severely on this transition. \( \Gamma_6^{e}P_{3/2} → \delta S_{1/2} \) may be uncertain by as much as 30%[5]. We estimate overall errors of \sim 45% in the measured EP rate coefficients.

Using the mean thermal velocity

\[ \tau = \left( \frac{8RT}{\pi} \right) \left( \frac{M_1 + M_2}{M_1M_2} \right)^{1/2} = 2.01 \times 10^{3} \text{T}^{1/2} \text{cm} \cdot \text{s}^{-1}. \]

(6)

We can calculate an average cross sections defined by \( \sigma_{nl_J'} = k_{nl_J'}/\tau \). The results are listed in Table 2.

Table 2. EP Rate Coefficients and Cross Sections for Rb(5P_J)+Cs(6P_3/2) → Rb(5S)+Cs(nl_J')

<table>
<thead>
<tr>
<th>J</th>
<th>nl_J'</th>
<th>Rate Coefficients (cm³s⁻¹)</th>
<th>Cross Sections (cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>4F_{5/2}</td>
<td>2.72 ± 1.2</td>
<td>\times 10^{-10}</td>
<td>7.2 ± 3.2</td>
</tr>
<tr>
<td>1/2</td>
<td>1.4 ± 0.6</td>
<td>\times 10^{-10}</td>
<td>3.8 ± 1.7</td>
</tr>
<tr>
<td>8S_{1/2}</td>
<td>14 ± 6</td>
<td>\times 10^{-10}</td>
<td>3.8 ± 1.7</td>
</tr>
<tr>
<td>4F_{7/2}</td>
<td>3.8 ± 1.7</td>
<td>\times 10^{-10}</td>
<td>10.2 ± 4.6</td>
</tr>
<tr>
<td>3/2</td>
<td>7.9 ± 3.6</td>
<td>\times 10^{-10}</td>
<td>21.1 ± 9.6</td>
</tr>
<tr>
<td>8S_{1/2}</td>
<td>7.9 ± 3.6</td>
<td>\times 10^{-11}</td>
<td>2.1 ± 1.0</td>
</tr>
</tbody>
</table>
The results of Table 2 show that Cs(8S) state is more effectively populated by Rb(5P\(_{1/2}\)) + Cs(6P\(_{3/2}\)) collisions than by Rb(5P\(_{3/2}\)) + Cs(6P\(_{3/2}\)) collisions. This can be explained strictly on the basis of energy deficits. Rb(5P\(_{1/2}\)) + Cs(6P\(_{3/2}\)) is more resonant than Rb(5P\(_{3/2}\)) + Cs(6P\(_{3/2}\)) with Cs(8S) (see Fig. 1(b)). For the Cs(4F) levels we find that Cs(4F\(_{5/2}\)) level is more strongly populated than Cs(4F\(_{7/2}\)) by Rb(5P\(_{1/2}\)) + Cs(6P\(_{3/2}\)) collisions while 4F\(_{7/2}\) is more strongly populated than 4F\(_{5/2}\) by Rb(5P\(_{3/2}\)) + Cs(6P\(_{3/2}\)) collisions. Thus the differences between Rb(5P\(_{1/2}\)) + Cs(6P\(_{3/2}\)) and Rb(5P\(_{3/2}\)) + Cs(6P\(_{3/2}\)) collisions may indicate that angular momentum propensity rules can play a role in the EP process.

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References