Possibility of buffer-gas cooling of paramagnetic carbon to ultracold temperatures

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We demonstrate that the rate constant for the collisional relaxation of $^3P_1$ in a buffer gas of He vanishes as the temperature decreases below 1 K. This suggests that the buffer-gas loading technique can be used for trapping excited paramagnetic $^3P_1$ atoms at ultracold temperatures.

Buffer-gas loading techniques have been developed recently for cooling and trapping paramagnetic atoms and molecules at ultracold temperatures [1–3]. The experiments are based on energy equilibration in the elastic collisions of atoms or molecules with the buffer gas atoms, usually He. The temperature of the buffer-gas varies between tens of mK and 1 K. The atoms cooled can be trapped in the magnetic field, the depth of the trap depending on the magnetic moment of the atoms. Although initially the atoms and molecules may be in excited levels, they relax quickly to the ground level due to inelastic collisions and in all experiments using buffer-gas cooling only atoms or molecules in the electronic ground states have been trapped. Thus, $^3P$ atoms with an inverted triplet of spin-orbit energy levels such as oxygen are regarded as possible candidates for experiments at ultracold temperatures, whereas $^3P$ atoms with the normal triplet energy level structure such as carbon were not considered for buffer-gas cooling because ground state $^3P_0$ atoms cannot be magnetically trapped. The trapping of carbon atoms at ultracold temperatures would, however, be highly desirable as it might allow for the precise analysis of their properties and lead to the generation of organic molecules at ultracold temperatures.

The spin-orbit energy levels of carbon are shown in Fig. 1. The purpose of the present article is to demonstrate that buffer-gas cooling of the metastable $^3P_1$ in He is possible and consequently carbon atoms in the $^3P_1$ state can be trapped at ultracold temperatures.

The dynamical calculations here are performed using a close coupling quantum mechanical approach that has been described in detail previously [4–7]. The potential for the interaction of a $^3P$ atom with He can be expanded in spherical harmonics as follows [8]:

$$V(R, \rho) = \sum_{\lambda} \frac{4\pi}{2\lambda + 1} \sum_{\mu} \sum_{i} V_\lambda(R, \rho_i) Y_{\lambda\mu}(\hat{R}) Y_{\mu\mu}^*(\hat{\rho}_i),$$  

(1)

where $\hat{\rho}$ denotes collectively the position vectors $\hat{\rho}_i$ of the $p$ electrons of the open-shell atom measured with respect to the nucleus of the atom, $\hat{R}$ is the vector joining the colliding particles and the symbols without carets denote the scalar quantities. For the present case $\lambda = 0$ and 2. The elements of the electrostatic (ES) interaction matrix coupling different spin-orbit states $^3P_j$ take the form

$$\langle jm | V(R, \rho) | j'm' \rangle = \langle jm | \sum_i V_0(R, \rho_i) | j'm' \rangle + \frac{4\pi}{5} \sum_{\mu} Y_{2\mu}(\hat{R}) \times \langle jm | \sum_i V_2(R, \rho_i) Y_{2\mu}^*(\hat{\rho}_i) | j'm' \rangle,$$

(2)

where $m$ is the projection of $j$. The expansion coefficients $V_0$ and $V_2$ are related to the interaction potentials of the molecule HeC in the ground $\Sigma$ and $\Pi$ states [9]. These potentials are computed with the state-of-the-art ab initio approach using the MOLPRO2000 suite of programs [10] and the unrestricted coupled cluster with single, double, and noniterated triple excitation level of theory based on the single reference restricted open-shell Hartree-Fock wave function.

![FIG. 1. Energy levels of carbon in the $^3P$ state](image)
and inelastic collisions of C$^\left(3P\right)_1$. The rate of the elastic energy transfer at \(T = 1\) K is almost two orders of magnitude smaller than the rate for the \(3\)P$\left(3\right)P_0\) relaxation.\[7\]

For the \(j = 0 \rightarrow j = 1\) transition the matrix elements in Eq. (2) vanish. The \(j = 0\) and \(j = 1\) states in the \(3P\) atoms are, however, coupled through the \(j = 2\) state by a sequence of ES and Coriolis couplings [6]. Except for collisions with zero total angular momentum, the \(j = 0 \rightarrow j = 1\) transition can therefore occur by a three-step mechanism involving an intermediate transition to the \(j = 2\) level and may be faster than the allowed ES coupled \(j = 0 \rightarrow j = 2\) and \(j = 1 \rightarrow j = 2\) transitions [18]. In the ultracold temperature limit collisions are dominated by s-wave scattering and the \(j = 0 \rightarrow j = 1\) relaxation in O$^\left(3\right)P_0$-He collisions is impossible [6]. The ultracold \(j = 1 \rightarrow j = 0\) relaxation in carbon is determined by the total angular momentum \(J = 1\) and it occurs through the coupling to the closed \(j = 2\) state that lies only 27 cm$^{-1}$ above the energy of the \(j = 1\) state (Fig. 1).

Figure 2 shows the calculated rate constants for the elastic and inelastic collisions of C$^\left(3\right)P_1$ with \(^3\)He atoms at temperatures between \(10^{-5}\) and 1 K. The rate for the \(j = 1 \rightarrow j = 0\) relaxation that would remove the trappable \(3\)P$\left(3\right)P_1$ carbon atoms is almost two orders of magnitude smaller than the rate of the elastic energy transfer at \(T = 1\) K and decreases quickly as the temperature goes to zero. The coupling between the \(j = 1\) and \(j = 0\) states is completely blocked at ultracold temperatures and the rate constant for the \(j = 1 \rightarrow j = 0\) transition does not obey the s-wave Wigner threshold law [19].
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