

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 C(³P₁) 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 C(³P₁) atoms at ultracold temperatures.

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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 ³P₀ 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 C(³P₁) in He is possible and consequently carbon atoms in the ³P₁ 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]:

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

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

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

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 Σ and Π 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,

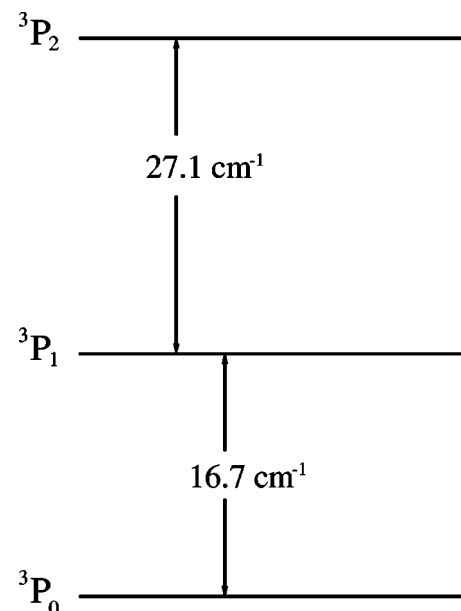


FIG. 1. Energy levels of carbon in the ³P state

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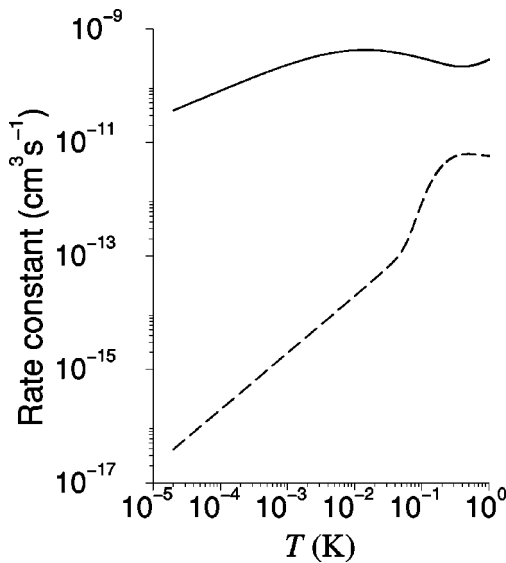


FIG. 2. Rate constants for elastic energy transfer (full curve) and the ${}^3P_1 \rightarrow {}^3P_0$ relaxation (broken curve) in $C({}^3P_1)$ - ${}^3\text{He}$ collisions.

R-UCCSD(T), as described in Refs. [11–13]. An extremely large correlation-consistent polarized valence basis set, aug-cc-pV5Z [14–16] augmented with an extended [3s3p2d2f1g] set of bond functions with the exponents [17], sp :0.94, 0.34, 0.12; df :0.64, 0.23; g :0.35 is used for the calculations. It has been demonstrated for the similar $O({}^3P)$ -He system [7] that the use of such potentials ensures an accurate description of ultracold collision dynamics. Formal expressions for the matrix elements (2) have been given previously [6]. All calculations of the present work are for collisions with ${}^3\text{He}$.

For the $j=0 \leftrightarrow j=1$ 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 $j=0 \leftrightarrow j=1$ 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 $j=0 \leftrightarrow j=2$ and $j=1 \leftrightarrow j=2$ transitions [18]. In the ultracold temperature limit collisions are dominated by s -wave scattering and the $j=0 \rightarrow j=1$ relaxation in $O({}^3P_0)$ -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 $C({}^3P_1)$ with ${}^3\text{He}$ atoms at temperatures between 10^{-5} and 1 K. The rate for the $j=1 \rightarrow j=0$ relaxation that would remove the trappable 3P_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].

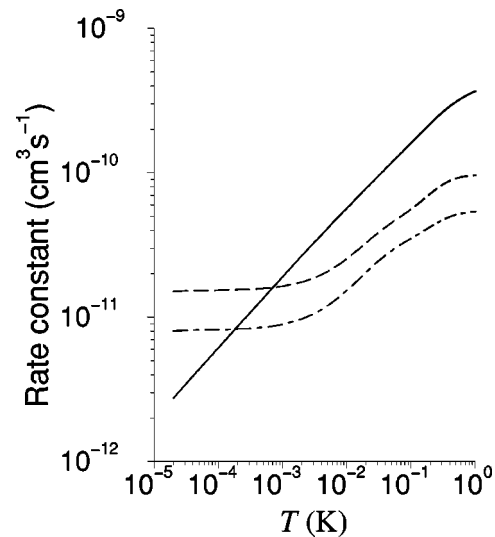


FIG. 3. Rate constants for $C({}^3P_2)$ - ${}^3\text{He}$ collisions: full curve, elastic energy transfer, broken curve, the ${}^3P_2 \rightarrow {}^3P_1$ transition, dot-dashed curve, the ${}^3P_2 \rightarrow {}^3P_0$ transition.

An analysis of partial wave cross sections for the $j=1 \rightarrow j=0$ transition shows that the cross section at total angular momentum $J=1$ decreases in the limit of vanishing collision energy $E \rightarrow 0$ as $E^{1/2}$ corresponding to the p -wave form of the Wigner law. The $J=2$ cross section has the energy dependence of the d -wave Wigner threshold law and the cross sections for all other J values have a threshold behavior corresponding to the orbital angular momentum $l=J$. The contribution from the initial channel with $l=J-1$ thus appears to be suppressed and the dynamics of ultracold inelastic He- $C({}^3P_1)$ collisions is modified by the three-step mechanism of the $j=1 \rightarrow j=0$ transition.

Figure 3 shows the rate constants for elastic and inelastic collisions of $C({}^3P_2)$ with ${}^3\text{He}$ over the same interval of temperatures. The rate constants for both inelastic transitions are very large and approach constant values as $T \rightarrow 0$, in accord with the Wigner law. The probability for the $j=2 \rightarrow j=1$ relaxation is several times larger than for the $j=2 \rightarrow j=0$ transition.

To conclude, we have shown that the inelastic relaxation of carbon in the metastable paramagnetic 3P_1 state due to collisions with He atoms is negligible in comparison with elastic energy transfer in the temperature interval 10^{-5} –1 K and the collisional relaxation of $C({}^3P_2)$ yields predominantly $C({}^3P_1)$. This suggests that the ultracold carbon atoms in the 3P_1 state can be produced efficiently using He buffer-gas cooling. The $j=1 \rightarrow j=0$ relaxation in $C({}^3P_1)$ -He collisions is an example of an inelastic transition that does not obey the s -wave Wigner threshold law due to the symmetry constraints imposed by the ES interaction.

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