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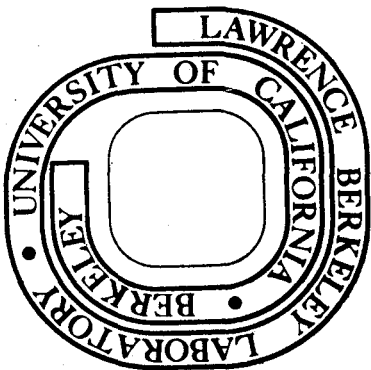
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William H. Miller

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Cooling of the 6 cm and 2 cm Doublets of Interstellar H₂CO by Collision:
An Accurate Quantum Mechanical Calculation

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ABSTRACT

Cooling of the 6 cm and 2 cm levels of interstellar H₂CO by a collisional pump is verified by a series of accurate quantum mechanical calculations. The $j = 3$ ortho doublet is found to be an integral part of the collisional pumping scheme.

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During the past few years, considerable interest has developed around observations of anomalous absorption in interstellar formaldehyde. This absorption is anomalous because it implies an excitation temperature for two rotational states lower than both the background radiation temperature ($\sim 2.7^\circ\text{K}$) and the expected kinetic temperature ($10\text{--}20^\circ\text{K}$). These observations are widespread throughout space and are seen in 1) the $1_{10} \leftarrow 1_{11}$ (6 cm) transition of H_2CO (Palmer et al. 1969; Dieter 1973; Minn and Greenberg 1973), 2) the $2_{11} \leftarrow 2_{12}$ (2 cm) transition of H_2CO (Evans, Cheung and Sloanaker 1970; Evans 1973; Evans et al. 1975) and 3) the $1_{10} \leftarrow 1_{11}$ transition of the isotope H_2^{13}CO (Evans et al. 1975). To obtain such low excitation temperatures requires a nonthermal cooling mechanism.

A number of pumping models have been proposed that involve transitions to higher rotational states of H_2CO and then radiative decay. The pump or force causing the excitations has been variously suggested as being due to collisions (Townes and Cheung 1969) or to radiation at millimeter (Solomon and Thaddeus 1970; Thaddeus 1972a), infrared (Litvak 1970), and ultraviolet (Oka 1970) wavelengths. Evans et al. (1975) have recently given a convincing discussion indicating that the collisional pump is the only model that accounts for all the observations and satisfies necessary criteria.

Since the collisional pump appears to be the key to understanding interstellar cooling of H_2CO , several workers have attempted to theoretically verify this model by determining the appropriate

rotational cross sections (Townes and Cheung 1969; Thaddeus 1972; Evans 1973; Augustin and Miller 1974; Evans et al. 1975; Evans 1975). These calculations suffer from either using approximate interaction potentials (hard or soft sphere), approximate dynamics (classical or semi-classical calculations) or inappropriate approximations (Born or sudden). In the present study, both the interaction potential and resulting cross sections and rates are determined using accurate quantum mechanical methods in order to obtain a reliable test of the collisional pumping model.

For these calculations, the most probable scatterer H_2 is replaced by He to reduce the scope of the computation. It is anticipated that the main conclusions of this study will not be seriously altered by this choice of scattering particle. To determine the interaction potential for the H_2CO -He system, we have solved the electronic Schrödinger equation including electron correlation (Garrison, Lester, and Schaefer 1975). The interaction is highly anisotropic and has a 35-40°K attraction arising mainly from dispersion effects. The well depth is believed reliable to ~ 20%.

For the dynamics, quantum mechanical cross sections were computed following an extension of the Arthurs and Dalgarno (1960) coupled-channel formalism to the scattering of an asymmetric rotor by an atom (Garrison, Lester, and Miller 1975). A total of 16 ortho H_2CO states were included in the coupled-channel expansion. Note that 4-8 states are energetically open at the scattering energies considered. Total inelastic cross sections were calculated at a

total of 12 energies for $20^\circ \leq E(^{\circ}\text{K}) \leq 95^\circ$, where E is the sum of the internal energy of H_2CO and the relative kinetic energy. Details of these calculations and results will be published elsewhere (Garrison, Lester, and Miller, 1975).

Inelastic cross sections were obtained for the lowest 8 ortho levels of H_2CO . (For convenience the energy levels are numbered in ascending order of energy (see Figure 1, Thaddeus 1972b), i.e., state 1 is 1_{11} , state 2 is 1_{10} , state 3 is 2_{12} , etc., thus the low excitation temperatures occur between states 1 and 2 and between states 3 and 4.) Many of the cross sections exhibited resonances at one of two energies, $E_{\text{total}} = 32.7^\circ\text{K}$ or 47.7°K . These energies correspond to the $j = 3$ and $j = 4$ ortho doublets, respectively. Boltzman averaged rate constants were determined from these cross sections and are given in Table I for a kinetic temperature of 15°K . Because the $j = 4$ cross sections are not determined at enough energies to form a Boltzman average, only rate constants for $j \leq 3$ are tabulated.

Using the computed rate constants, the equations of statistical equilibrium were solved for the lowest 8 energy levels. The background radiation temperature is assumed to be 2.7°K and the kinetic temperature is varied from 5 to 20°K . Plots of the excitation temperature as a function of He concentration are displayed in Figure 1. Cooling of both the 6 cm (T_{12}) and 2 cm (T_{34}) doublets is seen to occur for kinetic temperatures between 10 and 20°K , but not for 5°K .

Since resonance contributions to the cross sections can be large at the lowest energies considered, an attempt was made to estimate their importance on the cooling curves. The widths and heights were varied by a factor of two with no qualitative change in the cooling curves for He concentrations less than 10^5 molecules/cc. The quantitative change in the excitation temperatures, T_{12} and T_{34} , was found to be less than 0.2°K .

Having established that the 6 cm and 2 cm doublets of H_2CO are cooled by a collisional pump, the question of the relative importance of the various transitions remains to be fully elucidated. By varying the number of states used in the equations of statistical equilibrium, the effect of the different j doublets on the cooling can be assessed. Neglecting the $j = 4$ levels caused less than 0.2°K changes in the effective temperatures for He concentrations at which cooling occurs. Omission of the $j = 3$ levels, however, resulted in no cooling. Thus the $j = 3$ ortho doublet plays an integral role in the cooling of H_2CO . At low He concentrations ($\lesssim 10^5$ molecules/cc) radiative contributions are found to dominate collisional dipole allowed transitions so that rate constants k_{12} , k_{13} , k_{24} , k_{34} , k_{35} , k_{46} and k_{56} are of minor importance. Ratios of dipole forbidden transitions, e.g. k_{25}/k_{16} , are the indicators of cooling. The large ratio of $k_{25}/k_{16} \approx 6$ in Table I implies that transitions from the $j = 1$ to the $j = 3$ doublets are the prime determiners of the cooling.

For collisions of the isotopic homologue H_2^{13}CO the Born-Oppenheimer interaction potential is the same as before and all differences are contained in the dynamics treatment. They involve small changes in the center of mass of H_2CO , the reduced mass of the total system and the energy level spacing. These differences are expected to have little effect on the scattering cross sections. For these reasons it seems likely that the $j = 1$ doublet of H_2^{13}CO will also be cooled, in agreement with observations.

By a series of accurate quantum mechanical calculations, the collisional pump is confirmed as a cooling mechanism for the 6 cm ($j = 1$) and 2 cm ($j = 2$) doublets of ortho H_2CO . The $j = 3$ levels are found to be an integral part of the pumping scheme.

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Table I. Rate constants^a at $T_k = 15^\circ\text{K}$.

final state initial state	1_{11}	1_{10}	2_{12}	2_{11}	3_{13}	3_{12}
1_{11}	---	5.5	5.0	2.5	1.3	0.2
1_{10}	5.6	---	3.1	3.4	1.3	0.8
2_{12}	4.7	2.8	---	3.9	3.8	1.3
2_{11}	2.5	3.3	4.1	---	2.0	3.4
3_{13}	1.7	1.7	5.3	2.7	---	3.4
3_{12}	0.3	1.1	2.1	4.9	3.8	---

^a In units of 10^{-11} cc/molecule-sec.

Figure Captions

Figure 1: Excitation temperatures as a function of He density at selected kinetic temperatures.

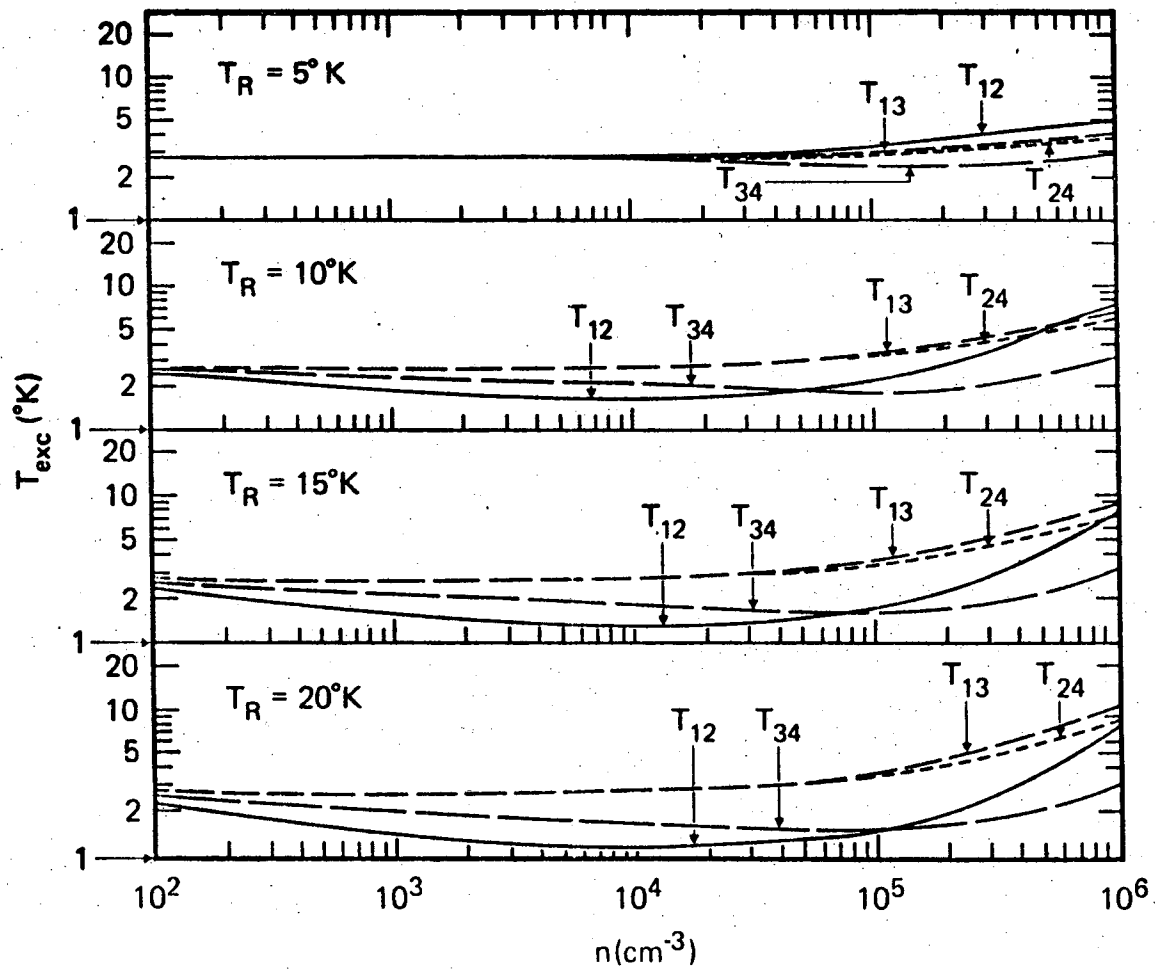


Fig. 1

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