Ultracold inelastic atomic collisions: Threshold relaxation of \( O(3P_0) \) by He

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Relaxation of \( O(3P_j=q) \) by collisions with He at ultracold temperatures is investigated, and it is illustrated that cross sections and rate constants for the \( j=0\rightarrow j=2 \) and \( j=0\rightarrow j=1 \) transitions exhibit qualitatively different threshold behaviors stemming from the different mechanisms of these nonadiabatic transitions. The results allow us to make the conclusion that, in general, rate constants for the nonadiabatic transitions governed by electrostatic couplings should obey Wigner’s threshold law for \( s \)-wave scattering approaching a finite value at \( T\rightarrow0 \) K, while cross sections and rate constants for the transitions determined by Coriolis couplings should vanish in the zero-temperature limit.

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Recent experimental success in the trapping and condensation of atoms and molecules at ultracold temperatures (see, e.g., Refs. [1–5]) stimulated theoretical investigations of inelastic molecular scattering near thresholds. In particular, Balakrishnan and co-workers [6–9], analyzed the low-energy limit of vibrational relaxation in collisions of diatomic molecules with atoms using the effective range theory and accurate quantum mechanical calculations. One of their main findings was that cross sections for vibrationally inelastic collisions at ultralow temperatures obey the \( 1/k \) Wigner threshold law (\( k \) is the wave vector for the collision) [10]. As a consequence, cross sections for vibrational relaxation tend to infinity as the collision energy (\( E_{\text{col}} \)) approaches zero, and the corresponding rate constant is finite in the zero temperature limit.

Recently, we analyzed the nonadiabatic quenching dynamics of \( O(3P_j) \) by rare gases at room temperature [11]. The intramultiplet relaxation of \( O(3P_0) \) has a peculiar mechanism with two possible pathways yielding the ground \( 3P_2 \) state and the first excited \( 3P_1 \) state of oxygen atoms. It was shown previously [12,13] that the \( j=0\rightarrow j=2 \) transition is induced by electrostatic coupling in the system, while the \( j=0\rightarrow j=1 \) transition, forbidden to first order, is determined by Coriolis coupling. We have illustrated [11] that the latter transition in fact proceeds through a complicated post-collision relaxation involving an interplay of electrostatically and Coriolis-induced nonadiabatic transitions. In the present work we extend quantum-mechanical calculations of Ref. [11] to investigate the relaxation dynamics of \( O(3P_0) \) by He atoms in the limit of low temperatures.

The dynamical calculations presented in this work are performed using the time-independent close-coupling quantum-mechanical approach described in Ref. [11]. All equations are solved in the body-fixed frame, and the resulting \( S \) matrix is transformed to the space-fixed representation of Arthurs and Dalgarno [14]. Cross sections and rate constants for inelastic scattering are then evaluated from the \( S \)-matrix elements using standard formulas. Corrections to Born-Oppenheimer approximation, which might be important for ultracold collisions, are ignored, and the standard model for spin-orbit interaction [11,12,15,16], parametrized by the non-relativistic O-He potentials from Ref. [17], is employed.

Figure 1 shows the dependence of the \( j=0\rightarrow j=2 \) and \( j=0\rightarrow j=1 \) cross sections on the collision energy \( E_{\text{col}} \). At energies between \( 1.5 \) and \( 6 \) cm\(^{-1} \), inelastic transitions are determined by scattering resonances. Separate calculations of elastic cross sections on the adiabatic potentials, as well as the similarity of resonance patterns for both inelastic transitions, demonstrate that the resonance enhancement is due to trapping of the system by the potential well of the entrance channel. The trapped system spends more time in the region where the electrostatic coupling between the \( \Omega=0 \) components of the \( j=0 \) and 2 levels is strong. At even lower energies, the behavior of cross sections for the two nonadiabatic transitions is different. The \( j=0\rightarrow j=2 \) cross section rises to infinity, while the \( j=0\rightarrow j=1 \) cross section vanishes. The inelastic cross sections at fixed orbital angular momenta \( l \) (partial opacity functions) are also shown in Fig. 1 for several partial waves making the biggest contribution to the resonant structure of the total cross section in the considered interval of energies. The energy dependence of all opacity functions except those for \( l=0 \) are similar for both electronic transitions. The \( l=0 \) partial cross section for the \( j=0\rightarrow j=2 \) transition rises to infinity, in agreement with Wigner’s threshold law for \( s \)-wave scattering [10]. The \( j=0\rightarrow j=1 \) transition at \( l=0 \) is, however, not possible, since there is no Coriolis coupling to drive the post-collision relaxation within the \( j=2 \) manifold of levels which is responsible for population of \( j=1 \) levels [11].

The different behavior of cross sections for the two relaxation pathways results in a different threshold behavior of rate constants for the \( j=0\rightarrow j=2 \) and \( j=0\rightarrow j=1 \) transitions (see Fig. 2). The rate constant for relaxation to the \( j=1 \) level tends to zero as temperature decreases, while the
relaxation rate to the \( j = 2 \) level approaches a finite value in the zero temperature limit. It is worth noting that, if the \( l = 0 \) cross section is excluded from the summation over partial waves, the two rate constants have similar but different by a temperature independent factor threshold behaviors.

To summarize, the relaxation of \( \text{O}(^3\mathrm{P}_0) \) presents a peculiar example in which two relaxation pathways exhibit qualitatively different behaviors in the low-temperature limit. The very transparent physics underlying this phenomenon allows us to conclude that, in general, cross sections for inelastic transitions induced by electrostatic couplings should follow the \( 1/k \) Wigner law for \( s \)-wave scattering at ultralow energies, whereas those determined by Coriolis couplings should vanish in the zero-velocity limit. This fact can be important not only for cooling atoms to ultralow temperatures, but also for equilibration kinetics in the interstellar medium.

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