Simulation of inelastic collisions and rotational cooling of ammonia in a 4 K helium buffer gas cell

C. J. Rennick[1], O. Schullian[2], J. Loreau[3], N. Vaeck[3], T. P. Softley[1]

[1] Department of Chemistry, Chemistry Research Laboratory, University of Oxford, Mansfield Road, Oxford OX1 3TA, United Kingdom.
[2] Laboratorium für Physikalische Chemie, ETH Zürich, CH-8093, Switzerland
[3] Service de Chimie Quantique et Photophysique, Université Libre de Bruxelles (ULB) CP 160/09, 50 av. F.D. Roosevelt, 1050 Brussels, Belgium

A buffer gas cell is a relatively straightforward approach to cooling a molecular species through elastic and inelastic collisions with a noble gas in a cell maintained at cryogenic temperatures. Such cells are used as a cold molecular beam source for experimental study of cold chemical reactions. Here, we consider the cooling of the internal (rotational) and translational degrees of freedom of ammonia as it passes through a 4 K helium buffer gas cell, before exiting through a small orifice.

The direct simulation Monte Carlo (DSMC) method is a widely-applied stochastic technique for integrating the Boltzmann equation for the evolution of the velocity distribution, considering elastic collisions. We have implemented a novel treatment of the inelastic collisions in a DSMC simulation by considering the state-to-state cross sections for the He-ammonia system. These cross sections are used to construct a probability matrix for rotational transitions, which is used in the collision step of our DSMC simulation to determine the post-collision rotational population distribution. Our model traces the collision-by-collision evolution of the rotational and kinetic energy distributions, and predicts the velocity and rotational state distribution of ammonia leaving the buffer gas cell, in excellent agreement with experimental measurements.