Abstract

In this thesis, collisions between nanometer-sized water clusters and surfaces are investigated, experimentally by cluster beam technology and theoretically by classical molecular dynamics (MD) simulations. The study is partly motivated by the need to better understand the function of rocket-borne detectors that are used in the characterization of *noctilucent clouds*, which sometimes appear in the atmosphere. The occurrence of these clouds is possibly linked to climate change.

The majority of the experiments were carried out on smooth graphite surfaces. Large clusters, consisting of several hundred molecules or more, were found to survive the collision with the surface. The scattering can be attributed to two fundamentally different inelastic scattering channels: *evaporation-mediated emission* and *direct scattering*. The evaporation-mediated emission is in operation at surface temperatures above ~700 K. The impacting cluster is heated by the surface, and as a result of this heating monomers and small fragments evaporate from the cluster. If the evaporation is vivid enough, a large fragment is pushed off the surface. The evaporating small fragments are partly accommodated to the temperature of the hot surface. MD simulations of equivalent collisions agree well with the results of the experimental investigations.

Direct scattering of clusters resembles the bouncing of macroscopic objects off surfaces, and is observed also at low surface temperatures. The clusters need to be larger than a few thousand molecules to scatter by this mechanism. MD simulations reveal that adhesion forces acting between the cluster and the surface control the outcome of the collision when the incident velocity in the direction normal to the surface plane is low. At high incident velocity, plastic deformation of the cluster is the controlling factor.

Experiments with the type of surfaces used in the rocket-based detection of noctilucent clouds were also performed. Clusters scatter directly also from these surfaces, and positively and negatively charged fragments are emitted during the collision. After long exposure times, the flux of positive cluster ions dominates the emission, and the ionization probability is proportional to the cross section of the scattered clusters. A transient current of negative polarity is, however, also present immediately after the exposure of the target surface to the beam.

The work performed in the thesis demonstrates the applicability of cluster science methods to investigations of ultrafine aerosol particles.

Keywords: water clusters, ice particles, cluster-surface interactions, collision dynamics, charge transfer, beam experiments, molecular dynamics simulations, noctilucent clouds

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