Optical Excitation and Decay Dynamics of Fullerenes

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Abstract

The dynamics of highly excited fullerenes have been studied experimentally. Attention has been paid to excitation processes, such as excitation of Rydberg states and excitation and lifetime of the triplet state in C_{60}. Both the latter processes where found to depend on the vibrational energy of C_{60}. The triplet lifetime decreases exponentially with increasing vibrational energy while excitation of Rydberg states requires a vibrational energy in the fullerene. Relaxation mechanisms were also investigated, in particular radiative cooling. For the large range of fullerenes measured the emissivity was measured to be $5{\cdot}10^{-4} - 15{\cdot}10^{-4}$. No difference in the radiation behaviour between empty fullerenes and fragments of the endohedral fullerene La@C_{82} was found. Moreover, clusters of fullerenes were observed to decay by fusion followed by sequential C_2 evaporation after femtosecond laser excitation.
In addition, the internal energy distribution obtained by molecules after multiphoton absorption from a laser was examined. For a realistic description of the laser and molecular beam the internal energy distribution is not well described by the commonly used Poisson distribution but rather follows a power law up to a cutoff proportional to the fluence.

Keywords: fullerenes, triplet state, radiative cooling, clusters of fullerenes, molecular fusion, Rydberg states, mass spectrometry, time of flight, endohedral fullerenes