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Intramolecular energy transfer, driving mechanisms, and reaction rates for collective motions of clusters[1]

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Conformational transitions of molecules, clusters, and biopolymers are typically large-amplitude collective motions that involve a large number of degrees of freedom in a coherent manner. One of the major challenges in modern molecular science is to understand the general mechanism for such collective motions. This talk highlights a novel dynamical mechanism for conformational transitions of atomic clusters in terms of intramolecular energy transfer and the driving forces for large-amplitude collective motions. First, we introduce a method of hyperspherical mode analysis, which generally classifies the (3n-6) internal modes of an arbitrary n-atom molecule into three gyration-radius modes, three twisting modes, and (3n-12) shearing modes. This hyperspherical mode analysis reveals that the gyration-radius modes are the primary collective modes that need to be activated in order for the system to achieve large-amplitude conformational transitions. Moreover, it illustrates how the twisting modes and the shearing modes critically initiate and trigger the conformational transitions by inducing the essential driving forces that activate the gyration-radius modes via mode coupling. Finally, we characterize this driving mechanism for conformational transitions from the viewpoint of the phase space geometry of the low-dimensional dynamical system of the gyration-radius modes. This reduced phase space geometry clearly accounts for the origin of non-statistical reaction rate processes of the clusters. The present method of hyperspherical mode analysis as well as the driving mechanism for collective motions could be widely applicable to conformational dynamics of complex molecular systems.

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