Stern-Gerlach deflection of magnetic molecules and clusters embedded in helium nanodroplets

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Magnetic beam deflection experiments on free molecules and clusters are complicated by the fact that their rotational and vibrational temperatures are often poorly known and sometimes ill-defined. Furthermore, internal excitation reduces their magnetic susceptibility. Helium nanodroplet embedding ensures well-defined cryogenic conditions for the dopants, as well as a high degree of field orientation of their magnetic moments. This orientation, analogous to that observed for electric deflections of polar dopants [1], makes it possible to use a Stern-Gerlach magnet [2] to obtain measurable beam deflections even for heavy nanodroplets carrying magnetic impurities.

Measurements on Fe(II) and Co(II) chloride molecules reveal strong paramagnetism of monomers (implying efficient spin thermalization and orbital angular momentum quenching), and antiferromagnetic ordering in dimers and trimers. Measurements on Na₂ dopants, in turn, exhibit in the most overt way possible the formation of high-spin alkali dimer complexes on nanodroplet surfaces.

Deflection measurements also demonstrate that even at fixed beam conditions, the average nanodroplet size carrying a specific dopant changes substantially as the pick-up density is varied. This has implications for characterization of droplet size statistics. An expression describing this effect was derived and verified by modeling of pick-up probabilities.

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