Rotational anisotropy decay as a tool to characterize the size distribution of doped rare-gas cluster beams

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Size-dependent effects are of particular interest in cluster physics. While precise size-dependent studies can be conducted for charged cluster species, determining the size of neutral clusters remains a challenge. Empirical models exist to predict the mean cluster size in a cluster beam based on expansion parameters [1]. However, in our experiments we are interested in the interaction between dopant molecules and cluster species. In this case, it is not clear how the size distribution of the doped clusters can be inferred from the empirical models. Effects such as evaporative cooling after doping, dopant pick-up and sticking probabilities can lead to differences in the size distribution between the initial cluster beam and the doped clusters.
We present a promising method to determine the doped cluster size distribution by measuring the decay of rotational anisotropy in a femtosecond pump-probe experiment (see Fig. 1a). The dephasing rate of the initially observed anisotropy is related to the doped cluster size distribution as different distributions lead to different ensembles of rotational frequencies. Challenges, like low sample density in the gas phase and dominating linear background signals are overcome by phase-modulation and lock-in detection [2]. Preliminary results suggest a high sensitivity on the cluster size of $\pm$ 50 atoms (see Fig. 1b). The approach is applicable to continuous and pulse beam expansion conditions.

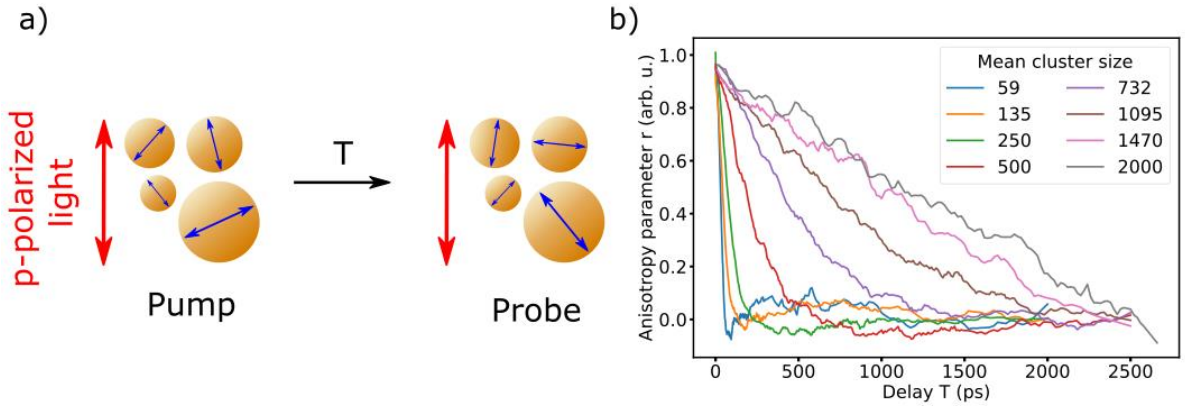

Fig 1. (a) Dephasing of the initially excited dipole moments, indicated by the blue arrows on the clusters. (b) Decay of the anisotropy parameter for argon clusters with a mean cluster size between 60 and 2000 atoms.

References:

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