Rotational Coherence Spectroscopy of Molecules in Helium Nanodroplets: Reconciling the Time and the Frequency Domains

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Alignment of OCS, CS_2, and I_2 molecules embedded in helium nanodroplets is measured as a function of time following rotational excitation by a nonresonant, weak ps laser pulse. The distinct peaks in the power spectra, obtained by Fourier analysis, are used to determine the rotational, B, and centrifugal distortion, D, constants. For OCS, B and D match the values known from IR spectroscopy. This result reconciles frequency-resolved spectroscopy and nonadiabatic laser-induced alignment dynamics. For CS_2 and I_2, the B and D constants are the first experimental results reported.

The alignment dynamics calculated from the gas-phase rotational Schrödinger equation, using the experimental in-droplet B and D values, agree in detail with the measurement for all three molecules.

The rotational spectroscopy technique for molecules in helium droplets introduced here should apply to a range of molecules and complexes. Also, it should allow studies of highly excited rotational states that are inaccessible with frequency-resolved spectroscopy.

[1] A. S. Chatterley, L. Christiansen, C. A. Schouder, A. V. Jørgensen, B. Shepperson, I. N. Cherepanov, G. Bighin, R. E. Zillich, M. Lemeshko, and H. Stapelfeldt, Phys. Rev. Lett. **125**, 013001 (2020).