Helium droplets offer new precision to single-molecule laser measurement

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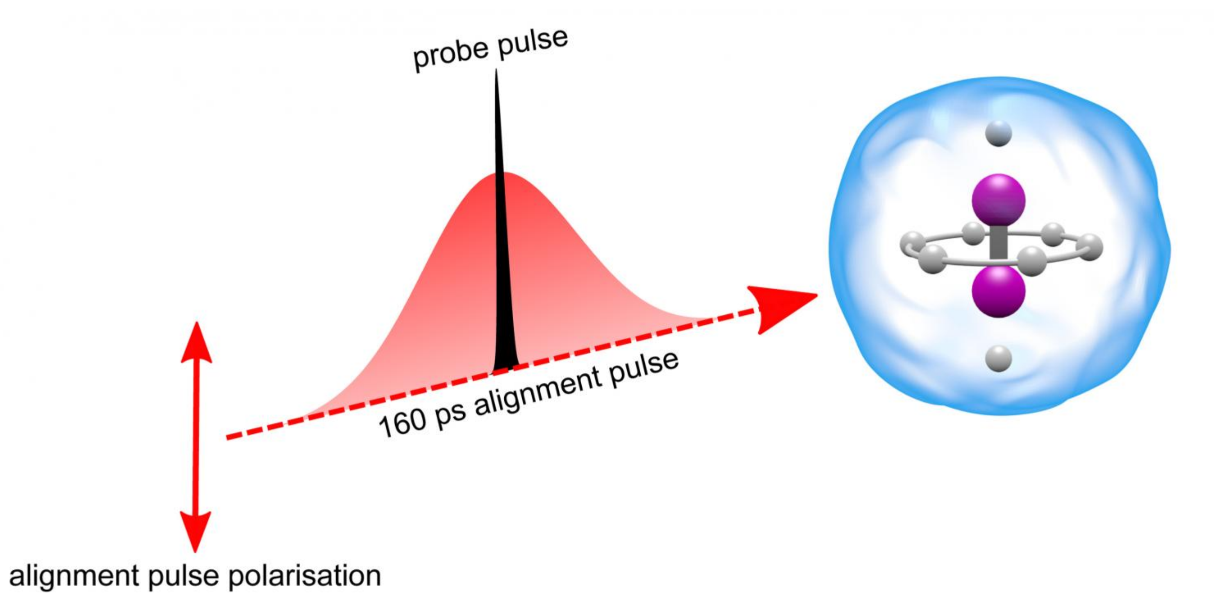

Schematic illustration of the alignment, induced by a 160 picosecond laser pulse (red), of an iodine molecule (purple) inside a helium droplet (blue). The iodine molecule is aligned vertically by the polarization direction of the alignment pulse, shown by the double-headed red arrow to the left. The degree of alignment is measured by a probe pulse (black) synchronized to the peak of the alignment pulse. Credit: Henrik Stapelfeldt, Aarhus University

Chemical reactions necessarily involve molecules coming together, and the way they interact can depend on how they are aligned relative to each other. By knowing and controlling the alignment of molecules, a great deal can be learned about how chemical reactions occur. This week in *The Journal of Chemical Physics*, scientists from Aarhus University in Denmark and the Institute of Science and Technology in Austria report a new technique for aligning molecules using lasers and very cold droplets of helium.

This new method aligns molecules more sharply than is possible for the essentially isolated molecules of those in the gas phase. This is due to the fact that a molecule embedded in a very cold droplet shares the same low temperature as the droplet itself, a mere 0.4 kelvins, or -272.75 degrees Celsius. It is only rarely possible to obtain such low temperatures for molecules in the gas phase, so this technique promises to open up a significant new regime for study.

The method utilizes a pair of laser pulses in what's called a pump-probe method. The first pulse aligns the single molecule once it has been deposited into a helium droplet. The second laser pulse, the probe pulse, is used to determine the alignment, blasting the molecule apart and separating it into ions. The ions fly off at specific angles and can be detected using a camera coupled to a computer.

"Being able to control the alignment of large molecules is no simple feat," Henrik Stapelfeldt of Aarhus University said, "because as molecules grow in size it becomes increasingly difficult to get them into the gas phase and cool them."

The investigators studied three systems: iodine (I2) molecules, which have a simple linear dumbbell shape, and two more complex molecules consisting of benzene rings with either iodine or bromine atoms attached to the ring. In all three cases, they achieved strong alignment of a single molecule embedded in a cold helium droplet with the two-pulse technique.

Because I2 has a simple linear shape, the investigators were better able to compare their experimental results to theoretical predictions. This revealed that the laser-induced alignment of molecules in helium droplets was essentially identical to that in the gas phase, as long as the alignment was done adiabatically, or gradually with respect to the molecules' responses.

To carry out adiabatic alignment, the first laser pulse is turned on more slowly than the inherent rotational period of the molecule being studied. This allows a freely rotating iodine molecule, say, to strongly align with the laser's polarization axis, in much the same way that a compass needle aligns with the magnetic field of the Earth.

Future studies will focus on aligning larger, more complex molecules in these cold helium droplets, allowing scientists to watch chemical reactions unfold in real time. Stapelfeldt explained that it may be possible to align molecules as large as proteins.

"Helium droplets offer unique possibilities," he said, "for building tailor-made molecular complexes, thus broadening the scope of systems that can be studied."

More information: "Strongly aligned molecules inside helium droplets in the near-adiabatic regime," *Journal of Chemical Physics* (2017). DOI: 10.1063/1.4983703

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