Isolating Water's Impact on Vibrations within DNA

Researchers in Berlin have found that water at the interface with DNA contributes prominently to subpicosecond structure fluctuations, and leaves hydrogen bonds between DNA and water intact

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By John Arnst

WASHINGTON, D.C., December 14, 2015 -- In a biological system, the ratio of water-to-non-water molecules, known as the hydration level, influences both the arrangement of biomolecules and the strength of the electric interactions that occur between biomolecules, free ions, and functional groups, which are groups of atoms within molecules that strongly influence the molecules' chemical properties. To isolate the contribution of water to the vibrational fluctuations that occur between DNA, bulk water, and the charged biomolecular interface between the two, researchers at the Max-Born Institute for Nonlinear Optics and Short Pulse Spectroscopy in Berlin have performed two-dimensional spectroscopic analyses on double-stranded DNA helices at different hydration levels.

The analysis gives insight into the way water and DNA interact, which could ultimately help scientists understand how biological systems function at the molecular level and what goes wrong when adverse conditions cause the systems to fail.

Two-dimensional infrared spectroscopy is a laser technique used to map correlated vibrations, the basic oscillatory motions of atoms, and their fluctuations into observable data.

The researchers used an amplified titanium-sapphire laser system to generate a sequence of four femtosecond infrared pulses in the low-frequency range that corresponds to the vibrational modes of DNA's sugar-phosphate backbones between 920 and 1120 cm^{-1}. They found that the spectra give evidence of both ultrafast structural fluctuations and a broadening of vibrational transitions, which reflects the structural disorder and variation of hydrogen bonding at the DNA-water interface.

"We were looking for probes which are most sensitive to dynamics at the [DNA-water] interface and noninvasive, leaving the structure of the interface unchanged," said Thomas Elsaesser, director of the institute and a professor of experimental physics at Humboldt University of Berlin. "We concluded that [DNA] backbone modes would be interesting candidates, as their elongations are at the interface and they should be sensitive to local electric fields."

Elsaesser and his colleagues at the Max-Born Institute detail their investigations this week in *Structural Dynamics,* from AIP Publishing.

Their current work builds on a seminal paper published in Nature in 2005 – with the collaboration of Dwayne Miller's group at the University of Toronto – which reported the first two-dimensional spectra of bulk water and established the basic time scales of the structural fluctuations that determine the lineshapes of vibrations.

For DNA, dehydration induces a transition from the traditional B-helix form to the A-helix form, the second most common shape. To determine the vibrational contribution of a system's hydration level, Elsaesser and his colleagues spectroscopically examined DNA strands at 0% humidity and 92% humidity, which correspond to around 2 and 20 water molecules per base pair, respectively.

By analyzing the two-dimensional spectral lineshapes, the researchers found that the hydrated DNA strands display structural fluctuations on a sub-picosecond time scale – less than a trillionth of a second – and that the structural disorder of local arrangements of water and DNA functional groups persists for time scales longer than 10 picoseconds, leaving water-DNA hydrogen bonds intact. Additionally, they found that although the arrangement of interfacial water molecules fluctuates at a slower rate compared to bulk water, it makes a substantial contribution to the sub-picosecond fluctuations, along with the low-frequency motions of the DNA helix.

They also noticed a pronounced coupling of the different, partly delocalized backbone modes. According to Elsaesser, this results in an energy transfer between the modes on a time scale of a few picoseconds.

Future work for Elsaesser and his colleagues includes extending their investigation toward longer natural DNA and RNA systems, such as DNA from salmon testes in a full water environment, as well as investigating the terahertz spectroscopy of low frequency motions and electric interactions.

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Ultrafast vibrational dynamics of the DNA backbone at different hydration levels mapped by two-dimensional infrared spectroscopy

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