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Nonlinear terahertz spectroscopy on liquid water

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Water is one of the most studied and least understood liquids[1]. While several anomalies in the thermodynamic, macroscopic properties of water are well documented[2], on the microscopic scale this special liquid is characterized by a dynamic, tetrahedral network of hydrogen bonded (HB) molecules rearranging on the picosecond timescale[3]. Thus, terahertz (THz) spectroscopy is uniquely suited to probe the dynamics of the HB water network.

The first results detailing the nonlinear response of water in the THz frequency range are recent, and the molecular interpretation is a matter of ongoing discussion. Tcypkin et al.[4,5], Ghalgaoui et al.[6], and Novelli et al.[7–10] all found that the third-order nonlinear response of water in the THz range has a similar magnitude, but proposed different explanations. The transient response of liquid water was assigned to cascaded second-order anharmonicity of the intramolecular O–H stretching modes[4]; to the field-induced irreversible ionization of water molecules[6]; or to the resonant excitation of molecular reorientations[8].

We performed a series of non-linear experiments at ~1 and 12.3 THz on liquid water at user facilities[7–10]. By comparing results obtained on a static sample and a free-flowing water jet at 12.3 THz, we were able to disentangle the distinct contributions by thermal, acoustic, and nonlinear optical effects[9]. Recently, we showed that the non-linear response of water at ~1 THz depends weakly on the temperature of the bulk liquid[10]. Based on these observations, we suggest that the THz fields could be reorienting the hydrogen-bonded water molecules in the liquid phase, as originally proposed in ref.[8].

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