Generation of Second Harmonic Light from a Low Number of Molecules in the Whispering Gallery Modes

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Unmarked sensitive detection of molecules is needed in many applications where a hidden substance must be identified. Typically, at least 1 billion molecules would be required to generate detectable light through an optical nonlinear process. By trapping the light in the whispering gallery modes of a high Q spherical micro-resonator we observed second harmonic light from less than 100 molecules. Quasi-phase matched second harmonic on the sphere surface was achieved by a periodic pattering of the molecular surface layer and pulse stretching to compensate temporal walk-off.

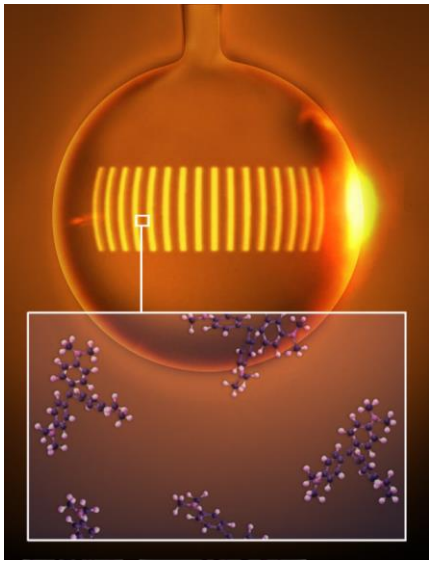

Figure 1 Picture of the microsphere and molecular periodic pattern.

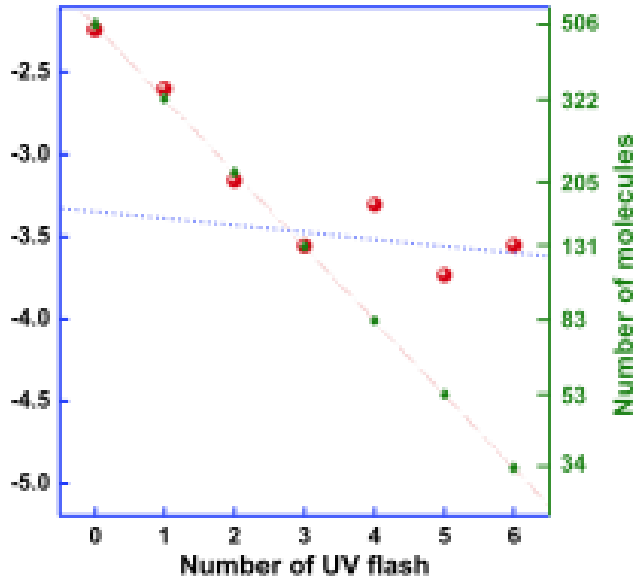

Figure 2 SH signal in a log scale (red spheres) as a function of the UV flash number used to destroy the NL molecules. The corresponding number of molecules (green dots) is estimated taking into account the initial surface concentration.

We report on the design and fabrication of a nonlinear spherical resonator to experimentally measure second harmonic generation (SHG) from molecules deposited on its surface. This generation requires phase matching in the whispering gallery modes, which we achieved by implementing a new procedure to periodically pattern with nanometric precision a molecular surface mono-layer, schematically shown in Figure 1. Temporal walk-off between the fundamental and SH pulses reduced the interacting length down to a very short distance. With radii of approximately 180 μm, pulse overlap at the perimeter of the sphere may be lost well before a single cavity round-trip is completed. To circumvent this problem the pump pulses were stretched using 140 m of an optical fiber loop to ensure permanent overlap. Using such type of walk-off

compensation allowed us to reduce the surface molecule concentration by four orders of magnitude. As shown in Figure 2, approximately between 50 to 100 molecules were needed to measure a change in the SH light [1, 2]. This result confirmed the relevant role played by such high Q micro-spheres. As indicated above, when molecules are deposited on a flat transparent substrate, typically one would need 1 billion molecules to obtain a measurable SHG signal. The possible application of nonlinear spherical micro-resonators in the detection of small molecules will be discussed.

[1] J. L. Dominguez-Juarez, G. Kozyreff, and J. Martorell, Nat. Commun. **2**, 254 (2011).
[2] G. Kozyreff, J. L. Dominguez-Juarez, and J. Martorell, Laser Photonics Rev. **5**, No. 6, 737–749 (2011).