



INSTITUTE SEMINAR

8TH JUNE, 2017 | 03:00 PM | FERMION HALL



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Thermally-driven nanoparticle assemblies as high-performing surface-enhanced-Raman-scattering substrates

ABSTRACT

Manufacturing products in the future will need new and improved engineering materials. The World Economic Forum The study of solid-vapor interface has strong implications in wide-ranging fields such as heterogeneous catalysis, analytical science, forensics, medical diagnostics, air-quality monitoring and international security. Probing vapors of solids at room temperature and pressure is highly desirable, but extremely challenging due to their low vapor pressure. To address this fundamental challenge, we demonstrate a route for real-time spectroscopic identification of a solid from its ambient vapor pressure. Vapor molecules present at the solid-vapor interface is used for ultra-sensitive (parts-per-billion), reliable (relative standard deviation < 3%) and rapid (< 70 s) sniffing of solids, including those with extremely low vapor pressure such as 2,4,6 trinitrotoluene ($\sim 10^{-9}$ atm). This is achieved through specially designed Soret-colloids acting as surface-enhanced-Raman-scattering (SERS) platforms. We observe that isolated nanoparticles form controllable and monodisperse assemblies (Soret colloids) under the influence of a thermal gradient. Thus, Soret colloids exhibit both intense and uniform electromagnetic 'hot-spots' and thus represent the synergistic combination of periodicity and uniformity of top-down, lithographically fabricated SERS platforms with the versatility of bottom-up, chemically-driven nanoparticle assemblies. Formation of such SCs is demonstrated to be a universally phenomenon across a variety of nanomaterials, with the kinetics strongly controlled by the dimension of the nanosystem and the thermal gradient. Thus, we overcome a fundamental dichotomy between ultrahigh, single-molecular level sensitivity and signal reliability for probing solid-vapor equilibrium. Importantly, SCs as SERS platforms present the only available route for ambient, real-time, quantifiable, single-molecular, spectroscopic analysis of both vapors and liquids, over wide concentration range (10^{-16} to 10^{-6} M).