Division of Synthetic Chemistry
–Advanced Inorganic Synthesis–

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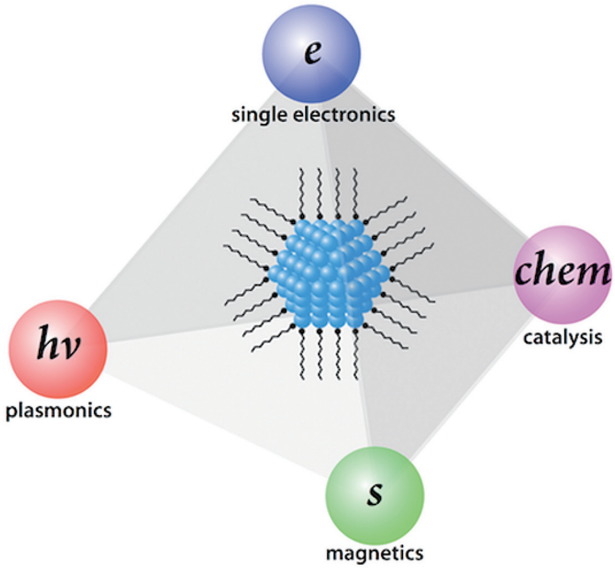
Prof PILENI, Marie-Paule University P&M Curie, France, 27 December

Scope of Research

We are focusing on the precise synthesis of inorganic nanoparticles by controlling the primary (size, shape, composition, etc.) and secondary (spatial arrangement) structures to tune their properties, such as electron confinement, carrier oscillation, spin, and catalysis. These high quality inorganic nanoparticles are applied to both high-performance nanodevices (single electron transistor, plasmon waveguide, nanocomposite magnet) and photo-energy conversion materials (overall water splitting, solar cell).



KEYWORDS

Inorganic Nanoparticles
Single Electronics
Plasmonics
Nanocomposite Magnet
Overall Water Splitting

Selected Publications

Saruyama, M.; So, Y. -G.; Kimoto, K.; Taguchi, S.; Kanemitsu, Y.; Teranishi, T., Spontaneous Formation of Wurtzite-CdS/Zincblende-CdTe Heterodimers through a Partial Anion Exchange Reaction, *J. Am. Chem. Soc.*, **133,** 17598-17601 (2011).
He, C. -L.; Chen, H. -Y.; Wang, C. -Y.; Lin, M. -H.; Mitsui, D.; Eguchi, M.; Teranishi, T.; Gwo, S., Far-Field Optical Imaging of a Linear Array of Coupled Gold Nanocubes at Visible Wavelengths: Direct Visualization of Dark Plasmon Modes, *ACS Nano*, **5,** 8223-8229 (2011).
Saruyama, M.; Kanehara, M.; Teranishi, T., Drastic Structural Transformation of Cadmium Chalcogenide Nanoparticles Using Chloride Ions and Surfactants, *J. Am. Chem. Soc.*, **132,** 3280-3282 (2010).
Kanehara, M.; Koike, H.; Yoshinaga, T.; Teranishi, T., Indium Tin Oxide Nanoparticles with Compositionally Tunable Surface Plasmon Resonance Frequencies in the Near IR Region, *J. Am. Chem. Soc.*, **131,** 17736-17737 (2009).
Li, C.; Sato, R.; Kanehara, M.; Zeng, H.; Bando, Y.; Teranishi, T., Controllable Polyol Synthesis of Uniform Palladium Icosahedra: Effect of Twinned Structure on Deformation of Crystalline Lattices, *Angew. Chem. Int. Ed.*, **48,** 6883-6887 (2009).

Charge Separation in Type-II CdS/CdTe Heterodimers Formed by Anion Exchange Reaction

Heterostructured nanoparticles (NPs), in which two or more distinct inorganic materials are connected together, are expected to provide new ways to manipulate wave functions, plasmon resonances, and spin. In semiconductor heterostructures, the choice of semiconductor materials allows control of the manner of confinement of the electron and hole wave functions in NPs. Type-II heterostructured NPs with a staggered alignment of band edges at the heterointerface can promote spatial charge separation of the electron and hole in different parts of the heterostructure for photocatalytic and photovoltaic applications.

The CdS/CdTe combination was chosen because this heterointerface forms a type-II band-edge alignment. The thermodynamically stable phases of CdS and CdTe crystals are wurtzite (*w*) and zinc blende (*zb*), respectively, and large strain at the heterointerface should induce spontaneous phase segregation. The anion exchange reaction proceeded smoothly to give anisotropically phase-segregated CdS/CdTe heterodimers with one heterointerface as an intermediate during formation of the completely exchanged product, CdTe NPs. Spontaneous formation of CdS/CdTe heterodimers took place because of the strain relaxation of the different crystallographic orientations of the *w*-CdS and *zb*-CdTe phases. As shown in Figure 1, atomic-resolution high-angle annular dark-field (HAADF) scanning transmission electron microscopy (STEM) observations revealed that the heterointerface was *w*-CdS (0001)/*zb*-CdTe ($\bar{1}\,\bar{1}\,\bar{1}$). Transient absorption spectroscopy demonstrated that photoinduced onedirectional charge separation occurred in CdS/CdTe heterodimers because of the type-II band-edge alignment with one heterointerface.

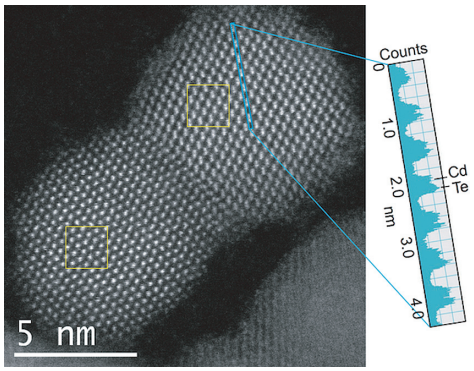


Figure 1. Atomic-resolution HAADF-STEM image of a single *w*-CdS/*zb*-CdTe heterodimer.

Dark Plasmon Propagation in Linear Array of Coupled Gold Nanocubes

Localized surface plasmon polaritons, which are referred to simply as localized surface plasmons, are collective electron oscillations in conductive NPs that are excited by incident light waves. Recently, the concept of optical nanoantennas based on plasmonic metal nanostructures has emerged as a promising route to realize optics and photonics far beyond the diffraction limit. In its inception, plasmonic nanoantennas composed of single metallic nanorods or nanoparticles were proposed as subwavelength receivers and/or transmitters of optical fields.

Recently, we have developed an alternative nanomanipulation technique for assembly of gold octahedron nanocrystal dimers with precisely controlled interoctahedron nanogaps. By studying the gold nanocube chains precisely assembled with tunable intercube separations, the fundamental plasmonic effects can be revealed in detail under well-prepared experimental settings, such as the number and spacing of nanocubes as well as the incident light direction and scattering light polarization. Using these chains, we were able to measure the dispersion relations of coupled plasmons with respect to the number of composing nanocubes and the intercube distance. In contrast to the existing results in the literature, we found that the plasmon coupling in gold nanocrystal structures could extend over a long spatial range (a few micrometers), resulting from the superior plasmonic properties of gold nanocubes, as shown in Figure 2. In addition, we confirmed that plasmonic waveguiding could only occur when the intercube distance is very small (much smaller than the nanocube feature size; under near-field plasmon coupling conditions).

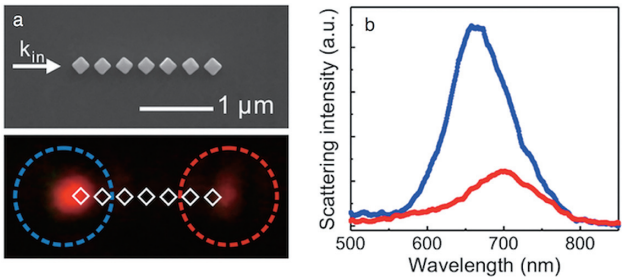


Figure 2. (a) FE-SEM image showing a chain composed of 7 uniformly spaced nanocubes (intercube tip-to-tip separation: 70±5 nm). (b) Scattering spectra acquired at both ends of the chain (regions marked by the dashed circles in corresponding colors in (a)).