NANO
2004

# SATELLITE WORKSHOP

## NANOPARTICLES

## SYNTHESIS, CHARACTERIZATION AND PROPERTIES

DUISBURG 25TH AND 26TH OF JUNE 2004

# BOOK OF ABSTRACTS

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**SFB 445** NANOPARTICLES FROM THE GASPHASE:
FORMATION, STRUCTURE, PROPERTIES

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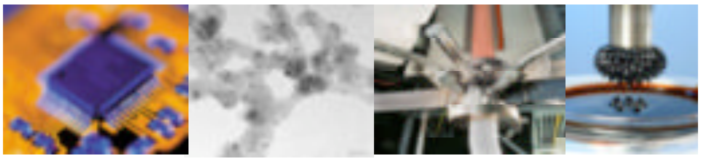

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NANO
2004

# SATELLITE WORKSHOP

## NANOPARTICLES

## SYNTHESIS, CHARACTERIZATION AND PROPERTIES

DUISBURG 25TH AND 26TH OF JUNE 2004

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**Local Organizing Committee:**

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# Friday, 25th of June

## 1. Properties

| | |
|---|---|
| 9:00 | Welcome |
| 9:15 | Ilona Kretzschmar (Yale): *Intrinsic Electron Conduction Mechanisms in Molecules* |
| 9:45 | coffee break / poster session |
| 10:30 | O. Hayden (Harvard): *Cadmium Sulphate Tripods for NanoLEDs* |
| 11:00 | A. Lorke (Duisburg): *Imaging of the Many-Particle Wave Function in Self-assembled Quantum Dots* |
| 11:30 | lunch break |
| 12:30 | M. Farle (Duisburg): *Surface Modification of Magnetic Nanoparticles by Plasma Etching* |
| 13:00 | poster / coffee break |
| 13:45 | R. H. Victora (Minneapolis): *Nanoparticles/grains for Magnetic Recording at 1 Tbit/inch* |
| 14:15 | coffee break / poster session |

## 2. Assembly

| | |
|---|---|
| 15:00 | J. Huskens (Twente): *Functionalized Nanoparticles in Surface Patterning and Nanofabrication* |
| 15:30 | H. O. Jacobs (Minneapolis): *Electrostatically Directed Site Specific Nanoparticle Deposition* |
| 16:00 | Ch. Mayer (Duisburg): *Spontaneous Embedment and Self-Organization of Nanoparticles in Phospholipid Multilayers* |
| 16:30 | coffee break / poster session |

## 3. Synthesis

| | |
|---|---|
| 17:15 | Sotiris E. Pratsinis (ETH Zürich): *Aerosol Synthesis of Dental Nanocomposites* |
| 17:45 | S. L. Girshick (Minneapolis): *Plasma Synthesis and Deposition of Nanoparticles for Coatings and for Microfabrication* |
| 18:15 | Transfer to Dinner |

**18:45 Dinner at *"Diebels im Hafen"***

## Saturday, 26th of June

### 3. Synthesis continued

| | |
|---|---|
| 9:00 | Welcome |
| 9:15 | U. Kortshagen (Minneapolis): *Plasma Synthesis of Highly Oriented Single Crystal Silicon Nano-Particles for Device Applications* |
| 9:45 | coffee break / poster |
| 10:30 | P. H. McMurry(Minneapolis): *Designing Aerodynamic Lenses for Focusing Nanoparticles* |
| 11:00 | E. Kruis: *Synthesis of Semiconducting Nanoparticles for Functional Devices* |
| 11:30 | lunch break |

### 4. Theory and Modelling

| | |
|---|---|
| 12:30 | S. C. Garrick (Minneapolis): *Direct Numerical Simulation of Nanoparticle Nucleation in Turbulent Flows* |
| 13:00 | P. Entel (Duisburg): *Structure and Magnetism in Transition Metal Clusters* |
| 13:30 | J. Roberts (Minneapolis): *Surface Chemistry at Size-Selected Nano-Aerosol Particles* |
| 14:00 | **concluding remarks, official end of workshop** |
| 14:30 | informal lab-tours (Duisburg) |

## Sunday, 27th of June: *Reunion in Xanten*
*"Rome – Minneapolis –Duisburg"*

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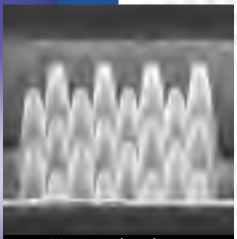
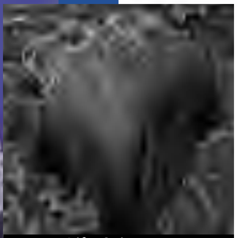
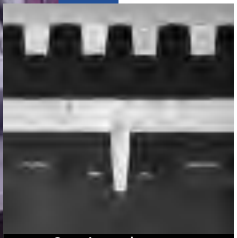


Nanotechnology

Materials Characterization



Life Sciences



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# ORAL PRESENTATIONS


PROPERTIES

# Intrinsic Electron Conduction Mechanisms in Molecules

Ilona Kretzschmar

Departments of Electrical Engineering and Applied Physics, Yale University

Electron devices containing molecules as the active region have been an active area of research over the last few years. This talk presents measurements in a variety of molecular systems to elucidate the transport mechanisms, specifically in self-assembled monolayers (SAMs) using nanometer scale devices. Detailed kinetic studies are necessary to distinguish between different conduction mechanisms; for example, in alkanes temperature-independent electron transport is observed, proving tunneling as the dominant conduction mechanism when defects are eliminated from the device structure. This is distinct from Langmuir-Blodgett films, where only defect or filamentary conduction has been observed. From the bias-dependence of $\beta$, a barrier height $\Phi_B$ of $1.39 \pm 0.01$ eV and a zero field decay coefficient $\beta_0$ of $0.79 \pm 0.01$ Å$^{-1}$ are determined for alkanethiols. Inelastic electron tunneling spectroscopy of the molecules in the junction exhibits well-defined modes of the molecules in the junction, and yield a measurement of the intrinsic linewidths of these modes. Deviation from this classic behavior for more complex molecule structures, and a comparison of the differences and pitfalls of various fabrication and characterization approaches, will be discussed.

# Cadmium Sulphide Tripods for NanoLEDs

Oliver Hayden[1,4], David C. Bell[2], Andrew Bardeen-Greytak[1],
Charles M. Lieber [1,3]

[1]Department of Chemistry and Chemical Biology,
[2]Center for Imaging and Mesoscale Structures, Department of Physics
[3]Division of Engineering and Applied Sciences, Harvard University, Cambridge, MA 02138
[4]Institute of Analytical Chemistry, University of Vienna, Währingerstrasse 38, A-1090 Vienna, Austria

The synthesis and optical properties of single zinc blende crystalline three-armed cadmium sulphide (CdS) nanowires (nano-tripods) using pulsed laser ablation is presented. The growth process and composition of these symmetrically-shaped CdS wires were thoroughly studied by (S)TEM, EDS and SEM from the nucleating point until the fully micrometer-long branched nanostructure. The initial nucleation stage of the structure begins with a single nano particle which then develops, through various stages, to a larger symmetric three armed structure (Figure 1). Interestingly the very central triangle of the structure that appears has a crystal orientation that distinctly belongs to only one of the three arms. The nano-tripods show a unique waveguiding pattern with end emission from the arms as well as from the centroid. PL results and nanoLEDs from tripods crossed with silicon nanowires will be presented and discussed.

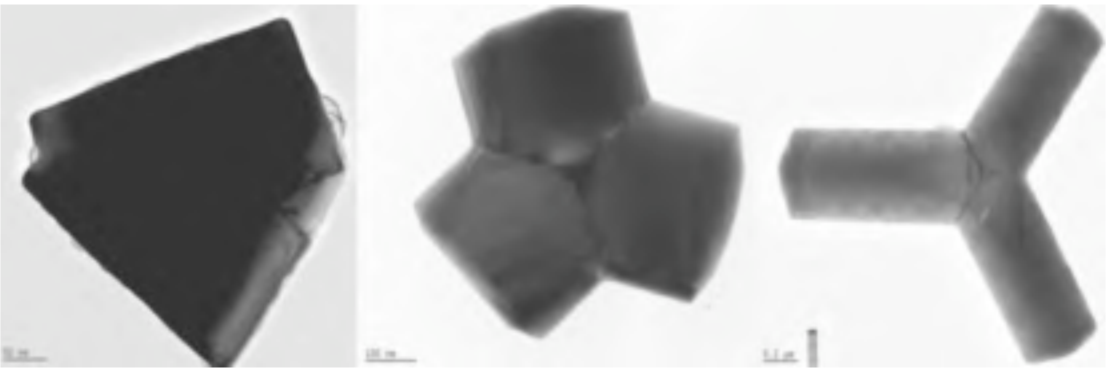

**Figure 1.** Stages of development during synthesis. From left to right, an early stage to final stage. The center image distinctly shows a triangular centroid that has a crystal orientation matching one of the arms.

# Imaging of the Many-Particle Wave Function in Self-Assembled Quantum Dots

Axel Lorke, Oliver Wibbelhoff

University Duisburg-Essen, Lotharstr. 1, ME 245, 47048 Duisburg, Germany

Self-assembled quantum dots have proved to be almost ideal model systems for studying electronic quantum effects in nanostructures. Properties like an energetic shell structure, Zeeman-splitting in high magnetic fields and the effect of exchange interaction have justified the term “artificial atoms” for these nanoscopic semiconductor structures. Contrary to real atoms, however, self-assembled quantum dots can be contacted electrically, they can be subjected to magnetic fields, which completely change their shell structure, and their electron number can be controlled *in-situ*.

Here, we will report on an additional feature that has recently been discovered in self-assembled InAs quantum dots: The possibility to image the electronic wave function inside the dots by magneto-tunneling experiments. Since the wave-function contains all essential information on the ground-state properties, this information is of great value for the understanding of the quantum nature of the electrons. The tunability of the electron number allows us to probe the wave function for 1 … 6 electrons per dot. We find that for the lowest state the data agrees well with a simple harmonic oscillator model. The higher states show clear evidence for electron-electron interactions and reveal an anisotropy of the confining potential. Furthermore, we observe a fine-structure in the electronic probability density which so far can not be explained within the common models for quantum dots.

# Surface Modification of Magnetic Nanoparticles by Plasma Etching

U. Wiedwald[1], K. Fauth[2], H.-G. Boyen[3], J. Lindner[1], M. Acet[1], M. Spasova[1], M. Farle[1]

[1]Institut für Physik, Universität Duisburg-Essen, 47048 Duisburg, Germany
[2]Physikalisches Institut, Universität Würzburg, 97074 Würzburg, Germany
[3]Abteilung Festkörperphysik, Universität Ulm, 89069 Ulm, Germany

Magnetic monodisperse particles like Co and $Fe_xPt_{1-x}$ with diameters on the order of 3 to 12 nm can be routinely prepared by organometallic synthesis. The magnetism of these particles is heavily influenced by unavoidable surface modifications due to the attachment of hydrocarbonic ligands and oxide formation. Even in the case of $Fe_x Pt_{1-x}$ particles [1] it has been unambiguously shown that Fe oxidation occurs under ambient conditions at the surface. To study the magnetism of the metallic core we demonstrate that a combination of hydrogen and oxygen plasma treatments under ultrahigh vacuum conditions (UHV) can be employed to completely remove the oxide and ligand layers from the surface. By combining para/ferromagnetic resonance techniques with x-ray magnetic circular dichroism and x-ray absorption spectroscopy we can identify for example the interface magnetism at the Co / CoO interface of a 12 nm Co particles covered with a 2 nm oxide shell [2]. The evolution of the ratio of the orbital-to-spin magnetic moment as a function of oxide-layer-removal shows that the magnetization of a 11 nm Co particle is fcc bulk like. 3 nm FePt particles can be cleaned and annealed in UHV on $Si_xO_{1-x}$ substrates without agglomeration showing enhanced magnetic anisotropies which may be of interest for future 10 Tb/inch data storage media.

Supported by Deutsche Forschungsgemeinschaft and EC – RTN “Magnetic nanoscale Particles” HPRN-CT-1999-00150.

[1] M. Ulmeanu, C. Antoniak, U. Wiedwald, M. Farle, Z. Frait, and S. Sun, Phys. Rev. B **69**, 054417 (2004)
[2] U. Wiedwald, M. Spasova, E. L. Salabas, M. Ulmeanu, M. Farle, Z. Frait, A. Fraile Rodriguez, D. Arvanitis, N. S. Sobal, M. Hilgendorff, and M. Giersig , Phys. Rev. B 68, 064424 (2003)

# Nanoparticles/grains for Magnetic Recording at 1 Tbit/inch$^2$

R. H. Victora, M. Patwari, X. Shen, J. Xue, and E. Yuan

Department of Electrical Engineering and Center for Micromagnetics and Information Technology (MINT), University of Minnesota.

As commercial hard disk drive densities approach 100 Gbits/inch$^2$, research has become increasingly focused on a density of 1 Tbit/inch$^2$. Bits with dimensions of 9nm x 60nm will need to be formed on thin films of thickness ~15 nm. In order to maintain accurate bit positioning (jitter) and low noise, the grains or particles within the film also need to be correspondingly smaller: 5nm diameter is a commonly accepted estimate. At these dimensions, thermal fluctuations can dominate and the material risks superparamagnetism (inadvertent switching of magnetization caused by thermal fluctuations.)

This talk will present results of our micromagnetic simulations for terabit recording systems. The recording medium is represented by Voronoi cells that include many of the complexities of experimental thin films. The equation of motion is taken to be the Landau-Lifshitz Gilbert equation. Thermal fluctuations are included through the Brown fluctuating vector formalism and a previously developed scaling approach that permits calculations for long time scales. We will discuss techniques used to ensure that grains are magnetically isolated, i.e., quantum mechanical exchange is broken. Techniques for combating superparamagnetism, such as composite media or 1 nm continuous coupling layers will be introduced and evaluated. Experimental implementation will be discussed.


ASSEMBLY

# Functionalized Nanoparticles in Surface Patterning and Nanofabrication

Jurriaan Huskens

University of Twente, MESA$^+$ Institute for Nanotechnology
Laboratory for Supramolecular Chemistry and Technology
P.O. Box 217, 7500 AE Enschede, The Netherlands
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Functionalized nanoparticles have powerful applications as intermediates between solution and surface chemistry and as tools for nanofabrication. Two main examples of these will be shown. The functionalization of nanoparticles with host sites will be described which allows: (i) the controlled aggregation with guest-functionalized dendrimers in solution, (ii) the specific adsorption onto patterned substrates, and (iii) the fabrication of larger architectures using the layer-by-layer methodology. The latter two employ so-called "molecular printboards" which are self-assembled monolayers with host recognition sites which allow the stable assembly of molecules and nanoparticles through multivalent host-guest interactions [1,2]. The second part will focus on the combination of (bottom-up) particle assembly and top-down surface structuring, in particular microcontact printing and nanoimprint lithography.

[1] J. Huskens, M. A. Deij, D. N. Reinhoudt, Angew. Chem. Int. Ed. 2002, **41**, 4467
[2] T. Auletta, B. Dordi, A. Mulder, A. Sartori, S. Onclin, C. M. Bruinink, C. A. Nijhuis, H. Beijleveld, M. Péter, H. Schönherr, G. J. Vancso, A. Casnati, R. Ungaro, B. J. Ravoo, J. Huskens, D. N. Reinhoudt, *Angew. Chem. Int. Ed.* 2004, *43*, 369.

# Electrostatically Directed Site Specific Nanoparticle Deposition

Chad R. Barry, Aaron M. Welle, Thomas J. Hatch, Heiko O. Jacobs*

Department of Electrical and Computer Engineering, University of Minnesota,
200 Union Street SE, Minneapolis, Minnesota 55455
*corresponding author, hjacobs@ece.umn.edu.

Nanoparticles are considered potential building blocks for the fabrication of future devices. This article reports on a new tool to direct the assembly of nanoparticles onto surfaces using electrostatic forces. We demonstrate directed self-assembly of nanoparticles onto charged surface areas (receptors) with a resolution of 100 nm. The charged based receptors required for this type of directed self-assembly were fabricated using a parallel method that employs a flexible, electrically conductive, electrode to charge a thin-film electret. The flexible electrode was brought in contact with a thin-film electret on an n-doped silicon substrate. The charge pattern was transferred into the thin-film electret by applying a voltage pulse between the conductive electrode and the silicon substrate. A new electrode design that is based on a 10 micrometer thick flexible membrane has been developed. Areas as large as one square centimeter were patterned with charge with 100 nm scale resolution in 10 seconds. Charge based receptors, 100 nm x 100 nm in size, contained ~100 elementary charges. These charge patterns attract nanoparticles. A liquid-phase assembly process where electrostatic forces compete with disordering forces due to ultrasonication has been developed to assemble nanoparticles onto charged based receptors in 10 seconds. A gas-phase assembly process has been developed that uses a transparent particle assembly module to direct and monitor the assembly of nanoparticles. Nanoparticles were generated using a tube furnace by evaporation and condensation. A process is also being developed to enable the patterning of any organic and inorganic material with sub 100 nm resolution. Currently, the electrostatically directed assembly of 10 – 100 nm sized metal and 30 nm – 3000 nm sized carbon particles is accomplished. Metal nanoparticles less than 100 nm in size can be printed at a resolution of 100 nm.

# Spontaneous Embedment and Self-Organization of Nanoparticles in Phospholipid Multilayers

Ch. Mayer[1], A. Terheiden[1], O. Dmitrieva[2], M. Acet[2], and B. Rellinghaus[3]

[1]Institut für Chemie. [2]Institut für Physik. [3]Institut für Verbrennung und Gasdynamik.
Universität Duisburg-Essen, D-47048 Duisburg, Germany.

FePt nanoparticles, synthesized and thermally annealed in the gas phase, are deposited onto Si substrates which are pre-coated with multilayers of amphiphilic phospholipids. With sufficient lateral molecular mobility, the multilayer serves as a reservoir for the spontaneous coating of the nanoparticles after deposition (Fig. 1). FePt nanoparticles are prepared by DC sputtering in Ar/He at $p = 0{,}5$ mbar and sintered in the gas phase at $T=600°C$. Under these conditions the obtained particles are monodisperse and of quasi-spherical morphology. The sintered FePt nanoparticles are deposited onto Si substrates which are pre-coated with multilayers of phospholipid molecules by the spin-coating technique. The resulting layer shows elongated inter-particle distances and a strong tendency to form regular particle patterns.


SYNTHESIS

# Aerosol Synthesis of Dental Nanocomposites

Sotiris E. Pratsinis

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Aerosol technology is attractive for synthesis of a number of high-value materials for its capacity to rapidly produce ultrapure particles with stable composition without liquid by-products [1]. In this lecture it will be presented how mixed oxide fillers are made with closely controlled characteristics so that can be used in manufacture of dental nanocomposites in close interaction with the corresponding dental material manufacturers. Capitalizing on our earlier success on synthesis of non-agglomerated, pure silica 70-100 nm in diameter for embedding it in dimethylacrylate polymer matrices [2,3], emphasis is placed on synthesis of silica-based mixed oxides focusing on the control of refractive index and radiopacity of the filler (nanostructured ceramic powders) and the final polymer-ceramic nanocomposite using flame-spray pyrolysis [4].

The filler production rate ranged from 6.7 – 100 g/h in a lab scale reactor. The effect of precursor, solvent and total metal concentration was studied by nitrogen adsorption, x-ray diffraction, light microscopy, HRTEM as well as the composite transparency within the polymer matrix of dimethylacrylate for dental restoration applications. Filler properties such as transparency, crystallinity and metal oxide dispersion in the $SiO_2$ matrix altered the composite's performance. The crystallinity and dispersion within the $SiO_2$ matrix affected the filler and composite transparency. Powders with identical specific surface area, refractive index and oxide-content showed a wide range of composite transparencies from 33 – 78 % depending on filler properties.

[1] SE Pratsinis, "Flame Aerosol Synthesis of Ceramic Powders" *Prog. Energy Combust. Sci.* **24**, 197 (1998).

[2] R. Mueller, H.K. Kammler, S.E. Pratsinis, A. Vidal, G. Beaucage, P. Burtscher "Non-agglomerated Dry Silica Nanoparticles", *Powder Technol.,* **140**, 40 (2004).

[3] G. Beaucage, H. Kammler, R. Mueller, R. Strobel, N. Agashe, S.E. Pratsinis, T. Narayanan "Probing the dynamics of nanoparticle growth in a flame by synchrotron radiation", *Nature Materials,* in press (2004).

[4] L. Maedler, W. Stark, S.E. Pratsinis, "Rapid Synthesis of Stable ZnO Quantum Dots", *J. Appl. Phys.,* **92**, 6537 (2002).

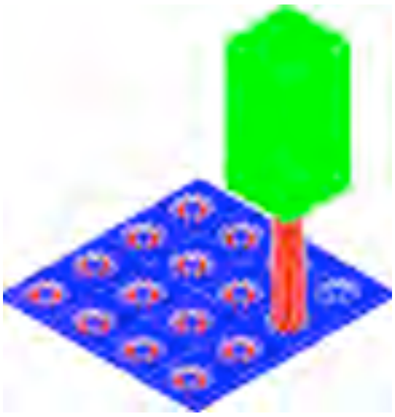
# Plasma Synthesis and Deposition of Nanoparticles for Coatings and for Microfabrication

Steven L. Girshick

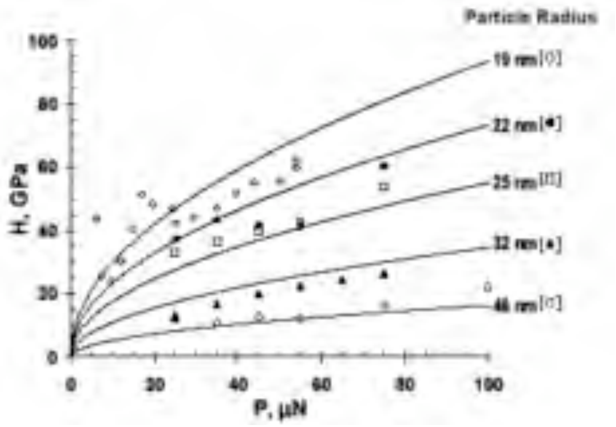
Dept. of Mechanical Engineering, University of Minnesota, Minneapolis, MN

Nanostructured films and coatings are promising candidates for applications requiring superior resistance to friction and wear. We have developed a process, known as hypersonic plasma particle deposition [1], in which nanoparticles of desired composition are synthesized in a thermal plasma that experiences a hypersonic expansion through a nozzle. Particles nucleate in the nozzle and are then accelerated in the expansion, until they impact a substrate to create a dense, nanostructured film. Films composed of silicon, titanium and their carbides have been deposited by this method. Alternatively, the substrate is replaced by a series of aerodynamic lenses, which focus the particles to a narrow beam [2]. A substrate is then rastered across the particle beam using a programmable translation stage, so as to accomplish microfabrication using nanoparticles. Current focus is on deposition of nanocomposites from the the Si-Ti-N system, which have been found to exhibit hardness comparable to diamond [3].

Several diagnostic methods are used to characterize both the particles and the film deposits. An in-situ aerosol sampling probe delivers particles from a specified reactor location both to a scanning mobility particle sizer, for on-line analysis of particle size and charge distributions, and to a small hypersonic impactor, which deposits particles onto TEM grids for off-line analysis. Deposited films are characterized by SEM, TEM, X-ray diffraction, and nanoindentation, and by a suite of diagnostics accompanying the TEM to analyze morphology, chemical and phase composition, at length scales ranging from the entire film to the atomic level. Particular emphasis is placed on the use of focused ion beam methods for TEM sample preparation. In recent work we have conducted nanoindentation studies of individual nanoparticles, and have shown that hardness increases dramatically as particle size decreases into the nanoscale regime [4].



Envisioned fabrication of MEMS gears using focused beam of nanonanoparticles.



Hardness of individual silicon particles of various initial sizes.[4] Each set of points corresponds to measured hardness vs. peak load for successive load-displacement curves for a given particle. The hardness of bulk silicon equals ~12 GPa.

This work is partially supported by NSF NIRT grant DMI-0103169.

[1] N. P. Rao, N. Tymiak, J. Blum, A. Neuman, H. J. Lee, S. L. Girshick, P. H. McMurry and J. Heberlein, *J. Aerosol Sci.* **29**, 707 (1998).
[2] F. Di Fonzo, A. Gidwani, M. H. Fan, D. Neumann, D. I. Iordanoglou, J. V. R. Heberlein, P. H. McMurry, S. L. Girshick, N. Tymiak, W. W. Gerberich and N. P. Rao, *Appl. Phys. Lett.* **77**, 910 (2000).
[3] S. Veprek, J. Vac. Sci. Technol. A **17**, 2401 (1999).
[4] W. W. Gerberich, W. M. Mook, C. R. Perrey, C. B. Carter, M. I. Baskes, R. Mukherjee, A. Gidwani, J. Heberlein, P. H. McMurry and S. L. Girshick, *J. Mech. Phys. Solids* **51**, 979 (2003).

# Plasma Synthesis of Highly Oriented Single Crystal Silicon Nanoparticles for Device Applications

A. Bapat[1], E. Thimsen[1], L. Mangolini[1], Ying Dong[2], C. R. Perrey[3], C. B. Carter[3], S.A.Campbell[1], and U. Kortshagen[1]

[1]Depar tment of Mechanical Engineering
[2]Department of Electrical and Computer Engineering
[3]Department of Chemical Engineering and Materials Science
The University of Minnesota, Minneapolis, MN 55455 USA

Nonthermal high density plasmas are a unique medium for the controlled synthesis of nanoparticles. Plasmas offer the same efficiency of direct gas-to-particle conversion as well as high process purity as other aerosol processing routes. Beyond that, however, nanoparticles in high density plasmas tend to be unipolarly charged thus effectively suppressing agglomeration. Hence extremely monodisperse particle size distributions can be achieved with as synthesized particles.

Crystalline semiconductor nanoparticles are of interest due to variety of electronic and opto-electronic applications such as, for instance, single-nanoparticle based vertical transistors and electro-luminescent devices. In this paper, we report experimental studies for synthesis and characterization of crystalline silicon nanoparticles using RF constricted-mode capacitive plasmas.

Monodisperse, nonagglomerated particles can be achieved in the size range of several tens of nanometers down to only a few nanometers. High-resolution TEM studies indicate high quality material with very low defect density. Larger particles of about 30 nm in size are deposited on metalized silicon wafers and Metal-Semiconductor-Metal (MSM) structures are fabricated. The conduction mechanism through the nanocrystals can be described by space charge limited conduction, where the space charge results from high levels of charge carrier injection rather than from trapped charges. Trap densities as low as one trapping state per particle are found, indicating that the produced nanocrystals are of high quality, virtually defect-free material and thus very suitable for device applications.

This work is supported by NSF under NIRT grant DMI-0304211 and grant CTS-9876224. AB, ET, LM, YD, SC, and UK acknowledge partial support by the MRSEC Program of the National Science Foundation under Award Number DMR-0212302. CRP, and CBC are supported by the 3M Heltzer Endowed Chair.

# Designing Aerodynamic Lenses for Focusing Nanoparticles

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Narrow particle beams with small divergence angles are desirable in many applications to enhance particle transport efficiency, improve measurement resolution or deposit micropatterns precisely at designed locations. Aerodynamic lenses [1, 2] have been widely used to generate such particle beams and are now used widely for aerosol mass spectrometers, etc. Current aerodynamic lenses can typically only focus particles larger than 20 nm. It is difficult to focus particle smaller than this due to their small inertia and high diffusivity. This talk will discuss design considerations for aerodynamic lenses that focus particles smaller than 20 nm.

To focus nanoparticles, the lens must operate at very low pressure. However, lower operating pressures enhance both diffusional losses and diffusional broadening of the particle beam. Required pumping capacities also increase as operating pressures decrease. We have found that there exists a maximum pressure under which particles can be optimally focused, while particle diffusion and pumping requirements are minimized. We will also discuss the effect of the carrier gas on particle focusing. It is shown that gases with lighter molecular weights permit higher operating pressures and assist focusing nanoparticles. Finally, we will present several case studies of designing aerodynamic lenses for focusing particles in the size range of 3 – 30 nm.

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# Synthesis of Semiconducting Nanoparticles for Functional Devices

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The synthesis of metal and semiconductor nanoparticles has attracted considerable interest due to their size-dependent structural, optical and electronic properties. An introduction is given into the specific characteristics of gas-phase synthesis methods of nanoparticles. The specific advantages such as high-temperature annealing into single crystals, the possibility of using size-fractionation techniques based on electrical mobility analysis and compatibility with techniques from microelectronics will be highlighted with examples from the synthesis of semiconductor nanoparticles.

A thin film synthesis technique with a high degree of control over the constituting particles has been developed. Well-defined nanoparticles can be prepared using a gas-phase condensation method. Alternatively, a laser ablation system allows the synthesis of compounds which do not evaporate congruently. Also, reactive laser ablation allows to synthesize compounds. As an example, GaN can be synthesized by ablation from a Ga target rod in a $N_2$ atmosphere. The resulting particles were sintered and crystallized in-flight. Size-selected nanoparticles ranging from 5 to 35 nm can produced to form a nanoparticle film by means of electrostatic precipitation or low-pressure impaction. The effect of in-flight chemical reactions, sintering and crystallization on the structure, size and size distribution of nanoparticles have been studied in detail. Care has to be taken that no evaporation takes place during the high-temperature in-flight treatment.

Zinc silicate doped with divalent Mn-ions is of great interest as it is commercially used as a green-emitting phosphor material. The luminescent properties of ZnO have received considerable attention due to its potential application in ultraviolet opto-electronic device and lasers operating at room temperature. Therefore, preparation of zinc silicate and zinc oxide with controlled stoichiometry and morphology is of great importance. Nanocrystalline zinc oxide and zinc silicate have been synthesized from single source molecular precursors by chemical vapour synthesis (CVS) process. The in-flight sintering of the size-classified ZnO particles has also been studied in order to get shape and size controlled ZnO for further applications in opto-electronic device.


THEORY AND MODELLING

# Direct Numerical Simulation of Nanoparticle Nucleation in Turbulent Flows

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In various processes, homogeneous nucleation occurring in turbulent flows is a key issue in determining the properties of the resulting aerosol. Examples can be found in nanoparticle generation (vapor-phase or combustion synthesis) as well as in atmospheric processes. As nucleation rates are very sensitive to small changes in conditions, it is clear that simulations must take into account the effects of turbulence-induced fluctuations. In this work, direct numerical simulation (DNS) of particle nucleation in shear flows is studied. DNS is advantageous in that it does not require assumptions of the structure of turbulence. Instead, the flow is solved directly in a spatially and temporally accurate/resolved manner. DNS is therefore a valuable tool for studying nucleation in turbulent flows as it does not require any restrictive assumptions of the mixing process (as well as providing data from which models may be constructed). Particularly, we analyze the onset of nucleation in planar jets. Homogeneous nucleation is considered to be the only transformation phenomenon affecting the particle population and is modeled using the nucleation model of Girshick & Chiu (1990). Results indicate that nucleation takes place in limited regions at the edges of the jet eddies where the laminar mixing of the two streams takes place. Depending on the mixing process (chemical, fluid, and thermal) and its state, different instantaneous nucleation rates are observed. As the jet progresses further, more mixing takes place and the nucleation rates increase. Lowering the co-flow velocity leads to more intensive mixing, more interfacial area between the streams and, as a result, significantly higher particle concentrations. Additionally some models are proposed for more affordable computations.

# Structure and Magnetism in Transition Metal Clusters

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We present a general overview about existing experimental and theoretical works on structure and magnetic properties of small iron clusters in order to get a deeper understanding of the quantum effects which govern the dependence of their physical and chemical properties in the nanometer range. We find that experimental information on structure and morphology of the clusters is very limited, while there is considerable amount of studies related to magnetic and electronic properties.

Calculations based on first principles have been performed by a number of groups using different approaches to describe exchange and correlation effects. There is, however, serious disagreement among them regarding important cluster properties like magnetic moments and ground state geometries even of the smallest possible entities. We show that this can be partly traced back to the degree of symmetry imposed during the calculations. From our calculations we find that energetic ground states are characterized by distorted shapes for all clusters considered.

# Surface Chemistry at Size-Selected Nano-Aerosol Particles

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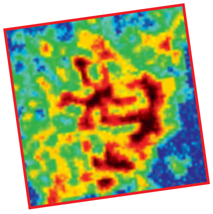
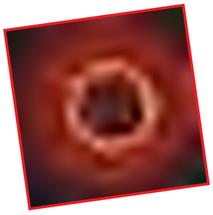
A method has been developed to conduct surface chemistry and extract surface kinetic rates from size-selected aerosol nanoparticles. The measurements encompass broad ranges of particle size, phase, and composition. Results will be presented on the growth and oxidation of soot nanoparticles of particle radius between 10 and 40 nm. Particles are investigated for changes in surface area and morphology using on-line instrumentation. Experiments emphasize two classes of reactive conditions: high temperature oxidation by $O_2$ and $NO_x$, and room temperature addition of hydrocarbons. We believe these to be the first measurements of soot oxidation and condensation kinetics that have been conducted on size-selected particles. The results are important because soot emission from combustion sources is dictated by the competing surfaces processes of growth and oxidation. More generally, the results represent one of the first kinetic and mechanistic studies of gas-phase nanoparticle reactivity.





# POSTER PRESENTATIONS


PROPERTIES

# Photoluminescence of GaAs Nanowhiskers Grown on Si Substrate

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Semiconductor nanowhiskers are a subject of intensive investigations since more than 35 years due to their unique growth features, interested physical properties and perspective device applications. Based on the Vapour-Liquid-Solid (VLS) growth mechanism [1], the fabrication of group-IV and III-V semiconductor nanowhiskers by chemical vapour deposition (CVD) and metal-organic vapour phase epitaxy (MOVPE) has been reported [2,3].

An important feature of the VLS growth mechanism is the predefinition of the growth area by the lateral size of a catalytic material, mostly Au. Using the nanoparticles as a catalytic centres the nanowhiskers with a diameter of a few nanometer can be realized [4]. Moreover, local growth on the nanometer scale area opens a possibility to fabricate the lattice strained one-dimensional semiconductor heterostructures. Successful realisation of some nanometer long defect-free InP/InAs/InP nanowhiskers with atomically sharp heterointerfaces [5] was already reported.

We investigated photoluminescence properties of GaAs nanowhiskers realised on Si substrate in VLS growth mode. As a catalyst the colloidal Au nanoparticles in the size range 5-100 nm were used. Growth procedure was performed in a MOVPE AIX 200 RF machine in a full non-gaseous source configuration at 400 °C. Scanning electron microscopy images show well defined GaAs whiskers with a length up to few μm. The photoluminescence spectra taken at 20 K demonstrate a strong peak at 820 nm corresponding to the band gap of GaAs. Intensity of the peak is comparable with this one measured on the reference sample grown on GaAs substrate. Further on, we perform detailed investigation of the photoluminescence behaviour in dependence on the temperature and excitation light intensity.

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# Size Dependent Optical and Electronic Properties of Si Nanoparticles

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Silicon nanoparticles (3.5-60 nm in diameter) were fabricated by pyrolysis of silane ($SiH_4$) in a low-pressure microwave reactor and investigated by transmission electron microscopy, Raman spectroscopy, and after deposition on a Au substrate, by Auger and electron energy loss spectroscopy. The particles consist of a Si core covered with a thin $SiO_x$ shell where the AES signal of oxygen correlates with the total area of the particle conglomerates. Both, first- and second-order Raman peaks of Si TO modes shift with decreasing particle size towards lower phonon energies. The increase in the intensity of the second-order modes may be due to an increase of the Raman cross-section since the electronic band gap increases with particle size and tends to resonant conditions. Likewise, the valence plasmon energy of Si exhibit a size-dependent shift from 16 to 17.7 eV with decreasing particle size.

# Exchange Interactions in Particulate Magnetic Nanostructures

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We present a combined simulation-experimental study of estimating the magnitude of exchange interactions in nanoparticulate systems. We have done in-situ ultra high vacuum x-ray magnetic circular dichroism experiments on a cobalt quasi-monolayer nanoparticulate system and used the data to obtain the hysteresis loops on 2 different directions of the applied field with respect to the plane of the sample.

SEM, TEM and AFM experiments are used to characterise the topography of the sample and to reveal the size distribution of the particles in the ensemble. The experimental sample is simulated using the observations and Landau-Lifshitz-Gilbert equation is assumed to describe the magnetic moments dynamic of each of the particles in the system. Dipole interactions between neighbour particles and exchange interactions between the particles which are observed to be in contact are taken into account.

Simulations are run to fit the two hysteresis loops having only two fit parameters, the average effective anisotropy of the particle and the exchange constant. The simulations produce unique values for these two parameters within small error bars. From the effective exchange interactions we estimate an average number of atoms per particle found in contact with neghbouring particles.

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# Magnetic Resonance on $Fe_xPt_{1-x}$ Nanoparticles

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Chemically disordered monodisperse 3nm $Fe_xPt_{1-x}$ nanoparticles with a protective ligand shell [1] were dispersed in hexane, a droplet of this solution was evaporated onto a substrate (quartz or TEM grid) and structurally and magnetically analyzed:
The ratio of orbital-to-spin magnetic moment averaged over the element-specific contributions of Fe and Pt has been measured for the $Fe_xPt_{1-x}$ nanoparticles at room temperature using the multi-frequency electron paramagnetic resonance (EPR) technique for different concentrations of Fe. From a detailed g-factor analysis [2] we determined that the ratio shows small changes. A comparison with calculated bulk values suggests that the induced orbital magnetic moment is antiparallel to the magnetic moment of the iron.
Magnetic resonance measurements on $Fe_{70}Pt_{30}$ at different temperatures yield a shift of the resonance field to much smaller values with decreasing temperature due to the increasing effective magnetization in the sample. Furthermore, both intensity and full-width-at-half-maximum have a maximum value corresponding to a mean blocking temperature of about 120K in the time window of the measurement (nanoseconds). By comparison with the blocking temperature we got from SQUID data (time window: 100s) the effective anisotropy constant for this composition can be calculated: it is $(8.4 \pm 0.9)\cdot 10^5 J/m^3$. This value is also found by fitting ZFC-magnetization-curves.

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# Multifrequency Magnetic Resonance of $CoPt_3$ – Nanoparticles

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$CoPt_3$ – nanoparticles, synthesized by the high-temperature organo-metallic route [1] and redispersed in a monomere (laurylmetaacrylate) followed by polymerisation, are investigated. Four different concentrations of $CoPt_3$-particles (0.08, 0.48, 0.8, 1.32 vol-%) and two different sizes (4 and 6.5 nm diameter) were prepared. Transmission electron microscopy (TEM) investigations show a chemically disordered fcc-phase with a slightly increased lattice constant a=0.389 nm (bulk $CoPt_3$ (fcc, chemically disordered): a=0.3829 nm; chemically ordered $L1_2$-phase: 0.3831 nm) [2]. Temperature dependence of field-cooled (100 Oe) / zero-field-cooled low-field susceptibility yield a blocking temperature of 12.5 Kelvin for the 4nm particles and 25 Kelvin for the 6.5nm particles. The size distribution, determined by TEM, shows a narrow standard deviation of $\sigma_d$=0.14 in the lognormal distribution. The percentage of surface atoms with respect to the total number of atoms per particle is 30% and 19% for the 4 nm and 6.5 nm diameters, respectively. Frequency dependent ferromagnetic resonance (FMR) measurements (4, 9.8, 24, 35, 48, 69.7 GHz ) at 300 K, i.e. in the superparamagnetic isotropic regime, yield a g-factor of 2.225 ±0.012 for the 4nm-particles and 2.30 ±0.1 for the 6nm-particles. This corresponds to an effective ratio of orbital-to-spin moment of $\mu_l/\mu_s$=0.113 ±0.006 (4nm) and 0.15 ±0.05 (6nm). The temperature dependence of the magnetic anisotropy is determined [4] for the 4nm particles. From the temperature dependence of the ferromagnetic resonance fields we find $K_2$=17.5 ±3.0$\mu$eV per $CoPt_3$-unit at 50 K down to 3.6 ±3.0 $\mu$eV per $CoPt_3$-unit at 350 K.

Supported by EC- contract HPRN-CT-1999-00150.

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ASSEMBLY

# Hygroscopicity and Volatility of Ultrafine Particles from Filtered Diesel Exhaust Aerosols

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Properties of particles emitted by a 1995 Caterpillar 3176 C-12 Diesel engine were studied with a nano-tandem differential mobility analyzer (TDMA) and a particle size distribution system (PSD). The engine was operated at constant speed (1500 rpm) and 32% load (570 N-m) using two ultra low sulfur Diesel fuels, 15 and 50 ppm. Engine exhaust was filtered through a Johnson Matthey Continuously Regenerating Trap.

Size distributions were measured with a nano-scanning mobility particle sizer (3nm<Dp<30nm) and a long-column scanning mobility particle sizer (20nm<Dp<200nm). The nano-TDMA includes a conditioner between two TSI model 3085 Nano DMA's. The conditioner is capable of either humidifying (10%<RH<90%) particles to study hygroscopic properties or heating (20C<T<150C) particles to study volatility properties.

Measurements indicated single mode distributions with average diameters of 6 nm (50 ppm S fuel) and 4 nm (15 ppm S fuel). The 70% reduction in fuel sulfur content eliminated over 80% of the total particle output. Hygroscopic growth factors of 1.15 were found for 4nm particles in both fuel types at 85% relative humidity. In the higher sulfur fuel, the relationship between particle size (4-15 nm) and growth factor (1.15-1.60) was measured at 85% humidity. Volatility measurements indicated no detectable nonvolatile components.

# The Use of Electric Nanocontact Lithography to Fabricate Patterns of Charge and Oxide on e-Beam Sensitive Resists

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We report on a programmable, reconfigurable, printing approach for parallel nanofabrication of two different types of structures: patterns of charge for nanoxerographic printing, and patterns of e-beam resist for nanofabrication. The developed tool is based on previous knowledge in the area of Conducting Scanning Probe Lithography, which uses a conducting probe to electrically expose and modify a surface. Our tool makes use of the same physical principles, however, instead of using a single electrical point contact, a conductive flexible electrode is used. This electrode is based on a flexible membrane that forms multiple electric contacts of different size and shape to the surface, exposing the entire surface to electrons and electric fields in a single step. The electrodes are used to expose e-beam sensitive resists to generate patterns of charge and patterns of topography with 100 nm resolution. Compared to the commonly used e-beam lithography, the electric nanocontact lithography provides a route to enable programmable, reconfigurable nanofabrication.

# A Nanoparticle-coated Nanocrystal-Gate for an InP-based Heterostructure Field-Effect Transistor

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Nanoparticle coated nanocrystals are fabricated by the Crystal Lattice Mediated Self-assembly method. While the core material is insulating the conductivity of the nanocrystals is provided by a coating with nanoparticles. Commercially available gold-nanoparticles are selected in order to provide a highly conductive element for a Schottky diode functionality of a junction to a semiconductor. This approach is used to fabricate the gate of an InP-based heterostructure field-effect transistor. The nanoparticle coated nanocrystal gate is deposited and aligned on the device structure by an electrostatic self-assembly via additional side electrodes. The gate voltage is applied to the gate using conductive-tip scanning force microscopy. The output characteristics of the nanoparticle coated nanocrystal gate FET with a gate width and length of 1 $\mu$m x 0.36 $\mu$m exhibits an extrinsic transconductance of 160 mS/mm.

# Synthesis of $SnO_2/TiO_2$ Mixed Oxide in a Doped Low Pressure Flame Reactor

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The importance of nanosized material in many contemporary applications is already known. However special properties such as superparamagnetism, semi-conducting and photocatalysis are unique to single metallic oxides. Recent advances in information technology, catalysis, and medicine often require materials that posses a combination of two or more of these properties. Nanocomposite materials present a possibility of having several unique properties within a compact material system. This "functionalization" of traditional single oxide system is the focus of many researches and the core competence of this investigation.
In this study, tetramethyl tin (TMT) and titanium tetra isopropoxide (TTIP) were diluted and sent through a low pressure premixed $H_2/O_2/Ar$ flame. The resulting $SnO_2/TiO_2$ metallic oxide was analyzed *in-situ* using PMS while BET, XRD, UV-VIS and TEM were performed *ex-situ*. Results show that the primary particle size of the mixed oxides can be controlled between $5\ \text{nm} \leq d_p \geq 12\ \text{nm}$ by varying experimental parameters. XRD and UV-VIS results indicate that a mixed oxide system was generated, and a defined tendency was observed with respect to the shift in the energy band gap of $TiO_2$. Anatase phase structure often associated with $TiO_2$ synthesis in premixed flame was not observed because $SnO_2$ acts like a "rutilizer" in this system. Band gap tuning opportunities thus exists by using mixed oxide systems.


SYNTHESIS

# Low Pressure Plasma Synthesis of Crystalline Silicon Nanoparticles

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Nonthermal plasmas are a uniquely suited for the controlled synthesis of nanoparticles. Nanoparticles in low-pressure plasmas tend to be negatively charged thus effectively suppressing agglomeration. Hence extremely monodisperse particle size distributions can be achieved with as synthesized particles.

In this poster, we discuss experimental studies of synthesis and characterization of crystalline silicon nanoparticles using different RF capacitive plasmas. The common feature of all plasmas is the deliberate excitation of a plasma instability. This instability leads to the formation of a high-density plasma filament, which favors the formation of crystalline particles. We describe the synthesis of crystalline silicon nanoparticles in a wide range of particle sizes. Particles are characterized by transmission electron microscopy, and by electrical and optical measurements.

This work is supported by NSF under NIRT grant DMI-0304211 and grant CTS-9876224 and under partial support by the MRSEC Program of the National Science Foundation under Award Number DMR-0212302.

# Thermal Plasma Chemical Vapor Deposition of Nanostructured Silicon Nitride/Carbon in a Triple Torch Reactor

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University of Minnesota

A triple torch reactor was used to deposit nanostructured silicon nitride/carbon films on silicon through thermal plasma chemical vapor deposition. Silicon tetrachloride vapor and acetylene/methane reactants were injected through a central injection probe and surrounding ring at flow rates of 0.01–0.04slm and 0.05–0.20slm, respectively. The deposition pressures ranged from 100–200Torr and the substrate temperatures from 900–1200°C. An argon-nitrogen plasma, with a gas flow ratio of ~20:1 was used to provide dissociated nitrogen to the multi-phase reactants.

The substrate temperature and reactant compositions were varied to correlate emission spectroscopy results with film composition and quality. The morphology of the composite films was determined by micro-XRD. The diffraction patterns of the crystalline films indicated hexagonal and tetragonal silicon nitride and hexagonal carbon. The bonding of the composite films was investigated with FTIR and XPS, indicating the presence of carbon-nitrogen bonds. The surface morphology was investigated by SEM, which depicted multifaceted structures deposited onto a nanostructured basis. Nanoindentation tests were conducted on the polished cross-sections of the films to determine the hardness and elastic modulus. It appears that the use of acetylene in the reactant mixture results in increased formation of NH radicals, as detected spectroscopically, and harder films.

# Plasma-Synthesis of Mixed-Phase Si Films: Nanocrystalline Si Inclusions in an Amorphous Si Matrix

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Mixed-phase Si thin films (a/nc-Si:H) have received considerable attention due to reports of superior electronic properties and an improved resistance to light-induced defect formation. In this study, a/nc-Si:H thin films are synthesized via radio-frequency plasma enhanced chemical vapor deposition with helium and hydrogen diluted silane. The plasma conditions are chosen to simultaneously deposit both Si nanocrystallites and an amorphous Si matrix. This structure has been confirmed by transmission electron microscopy (TEM) and high-resolution TEM (HRTEM). However, the formation of the nanocrystallites observed in the film matrix is not well understood. Nanocrystallite formation in the plasma is elucidated from several factors: the change in plasma impedance under a/nc-Si:H conditions, the increase in a/nc-Si:H film surface roughness, and the dependence of the deposited materials' structure on thermophoresis.

SST has been supported by NSF IGERT grant DGE-0114372. SST and UK are also supported by DOE grant DE-FG02-00ER54583 and NSF MRSEC DMR-0212302. CRP, JMD and CBC are supported by the 3M Heltzer Endowed Chair.

# Hypersonic plasma particle deposition of Si-Ti-N nanoparticle films

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Nanocomposite Si-Ti-N films are of interest for superhard coating applications. Hypersonic plasma particle deposition was used to deposit such films directly from nanoparticles. Particles were nucleated by injecting chloride vapors of silicon and titanium, together with ammonia, into an argon-hydrogen or argon-nitrogen plasma generated by a direct-current arc, and then expanding the plasma through a nozzle from about 55 kPa to about 270 Pa. Particles were deposited on a substrate by hypersonic impaction to form a film, or were collected by a sampling probe for measurement by a scanning electrical mobility spectrometer.

Films of 20-50 micron thickness were deposited on molybdenum substrates at rates of 2-10 $\mu$m/min, and substrate temperatures ranging from 250 $^0$C to 850 $^0$C. Microstructural characterization of the films was performed using standard methods. Hardness of as-deposited films was evaluated by nanoindentation equaled 22-26 GPa.

In-situ particle measurements showed that particle size distributions peak around 10 nm under typical operating conditions. The effects of particle residence time and partial pressure of nitrogen in the reactor on size distributions are under investigation. Particle diagnostics provide insight into the mechanism of particle formation and growth, and enables the optimization of operating conditions to achieve desired film properties.

# Fabrication of Magnetic Hollow Microspheres

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Hollow capsules are an important class of materials that can be obtained from precursor core-shell particles by direct removal of the core material using a variety of chemical and physical methods. We demonstrate a new method for the fabrication of *magnetic hollow spheres with tunable shell properties* from composite colloidal core-shell microspheres [1]. The composite core-shell particles were prepared by coating of anionic polystyrene spheres of 640 nm diameters with a 12 nm magnetite ($Fe_3O_4$) nanoparticle shell alternately absorbed with polyelectrolyte from aqueous solutions utilizing electrostatic interactions [2]. The thickness of the composite multilayer shell can be tuned between 20 and 180 nm by the number of absorption cycles. Hollow, intact magnetic spheres were obtained by oxygen plasma calcination of the composite particles deposited on SiO or GaAs substrates. As demonstrated by transmission and scanning electron microscopy, after plasma treatment the particles remain spherical and intact. Removal of the polystyrene core was confirmed by X-ray Energy Dispersive Spectroscopy (XEDS). These nanoengineered inorganic capsules may find applications in bio- and medical technologies as well as tunable photonic crystals.

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[1] E. L. Bizdoaca, M. Spasova, M. Farle, M. Hilgendorff, L. M. Liz-Marzan, F. Caruso, J. Vac. Sci. Technol. A21 (2003) 1515
[2] F. Caruso, M. Spasova, A. Susha, M. Giersig, R.A. Caruso, *Chem. Mater.* 13, 109 (2001).

# Water-based Magnetic Nanoparticles

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The organometallic synthesis of monodisperse magnetic nanoparticles with sizes ranging from 2 to 20 nm in organic solvents is of significant importance dor potential applications, because of its simplicity and control. For biological applications such particles need to be transferred to a non-toxic agent like water. Following a basic procedure [1] we demonstrate the synthesis of 3 nm $Fe_xPt_{1-x}$ ($0.3 \leq x \leq 0.7$) nanoparticles in non-polar solvents. A layer of surfactant molecules is essential to prevent the nanoparticles from aggregation. This organic layer, which causes the nanoparticles to be immiscible in the aqueous solution is replaced by another type of surfactants, tetramethylammonium hydroxide, and water-based magnetic $Fe_xPt_{1-x}$ dispersions are obtained.

In addition to alloys, core-shell structured $Co@SiO_2$ nanoparticles (approx. 25-200 nm diameter) were synthesized and studied. Cobalt colloids have been homogeneously coated with silica using the silane coupling agent (3-aminopropyl)-trimethoxysilane as a primer to render the cobalt surface vitreophilic. The thickness of the silica layer can be controlled by the amount of tetraethoxysilane used in the procedure and the particles can be transferred into polar and non-polar solvents.

The reversed core-shell structure can also be created using silica spheres as cores and depositing a thin shell of magnetic Co/CoO material.

Supported by EC contract HPRN-CT-1999-00150.

[1] S. Sun *et al.*, *Science 287* (2000) 1989

# In-Situ Analysis of Chemical Vapour Synthesis Processes in Hot Wall Reactors by Aerosol-Mass-Spectrometry

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Silicon Carbide nanoparticles are synthesized in a hot wall reactor by the thermal decomposition of tetramethylsilane (TMS). An Aerosol-Mass-Spectrometer is used to measure particle size distributions and molecular reaction products in a hot wall reactor. The sampling of the aerosol from the hot zone of the reactor is realised using a modified furnace, which is directly attached to the Aerosol Mass Spectrometer.
At about 900 K TMS starts to decompose and Carbosilanes with two [-Si–C-] units are identified as “starting spezies” in the synthesis of SiC from TMS. With increasing temperatures particles are formed and grow by coagulation processes. At higher temperatures sintering of the particles becomes dominant. Although the particle mass reduces slightly due to a smaller residence time at higher temperatures in the reactor, the particle velocity in the molecular beam of the AMS decreases significantly. Simulations from a monodispers coagulation-sintering model predict an increase of the sintering-rate in the observed temperature range. Ex-situ measurements comparing gas adsorption (BET) and X-ray diffraction (Scherrer equation) are consistent with an increasing sintering-rate. A simple model of Dahneke [1] is used to compare the particle velocity in a molecular beam as a function of particle mass. The significant difference in the particle velocity can be explained by a change in the particle shape factor ($\kappa$) due to sintering.

[1] B. E. Dahneke and Y. S. Cheng, J. Aerosol Sci. **10** (1979), 257-274

# Characterization of a Thermal Plasma Nanostructured Cubic Boron Nitride Deposition Process

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Cubic boron nitride (cBN) is a superhard material that has great potential for many applications, including wear-resistant coatings and opto-electronic devices. Although the deposition mechanisms of cBN are complex and not clearly understood for every deposition process, the necessity of energetic ion bombardment in cBN synthesis has been widely accepted. As thermal plasma deposition systems have great potential for nanostructured cBN deposition due to high energy densities, potential for rapid, uniform deposition over large areas, and large ion fluxes, quantitative data on critical ion bombardment parameters for cBN deposition in these systems is quite useful.

We have synthesized nanostructured cubic boron nitride films via thermal plasma chemical vapor deposition using a supersonic DC arcjet plasma. Ion bombardment is accomplished by electrically biasing the substrate holder. The films have been characterized by Fourier transform infrared spectroscopy, micro x-ray diffraction, and scanning electron microscopy. To further understand the effect of substrate bias in our system, an electric probe is used to measure the plasma parameters at different positions in the plasma jet under different biasing conditions. The results of the film characterization, and preliminary results from the electric probe measurements will be presented.

# HRTEM Investigations and Contrast Simulations of the Crystal Structure of FePt Nanoparticles from the Gas Phase

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We present the structural characterisation of FePt nanoparticles using high resolution transmission electron microscopy (HRTEM). The nanoparticles are prepared by inert gas condensation and subsequent sintering in the gas phase. The morphology and crystal structure of the obtained particles depend strongly on the gas pressure p and the sintering temperature $T_s$. Whereas particles prepared under p=0,5 mbar and with $T_s$< 600°C are monodisperse and of icosahedral structure, higher pressures and sintering temperatures lead to the formation of larger, polydisperse and increasingly $L1_0$-ordered face-centered tetragonal (fct) FePt nanoparticles. There are two interesting experimental findings which are analysed in details by HRTEM investigations combined with contrast simulations. First, at intermediate pressures and sintering temperatures many particles show still icosahedral signatures, but they are deformed as compared to regular icosahedra. The structure of these particles has been clarified. Second, many of the particles, which are prepared under p>1,5 mbar and with $T_s$=1000°C, show the $L1_0$-ordering in HRTEM-imaging, but not in electron diffraction patterns. To determine the amount of $L1_0$-ordered particles and the degree of order, we carried out contrast simulations and a statistical evaluation of the HRTEM images.

# Surface Oxidation of Size-Selected, Aerosolized Soot Nanoparticles

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The kinetics of reaction between soot nanoparticles and simple oxidants, including molecular oxygen, were studied by tandem differential mobility analysis. The particles were extracted from a laminar diffusion flame fueled by ethene and air. Reactions were studied at atmospheric pressure over broad ranges of temperature (room temperature - 1100 °C), initial particle size (30 nm mobility diameter), and oxygen partial pressure (0 -1 atm). Measurements as a function of oxygen partial pressure establish that the oxidation kinetics are not first-order in oxygen pressure ($P_{O2}$). Rather, the oxidation rate increases rapidly and linearly with $P_{O2}$ between 0 and 0.05 atm, and then more slowly with $P_{O2}$ between 0.05 and 1 atm. Kinetic modeling is consistent with an oxidation pathway involving two kinetically distinct sites for surface oxidation, $S_A$ and $S_B$, as shown below:

$$S_A + O_2 \xrightarrow{k_a} S_A + 2CO$$

$$S_B + O_2 \xrightarrow{k_b} S_A + 2CO$$

$$S_A \rightleftharpoons S_B$$

$$k_{a\_b}$$

$$k_{b\_a}$$

$$k_i = A_i T^{1/2} \exp[-E_{ai}/RT]$$

The scheme also allows for the thermally activated interconversion of $S_A$ and $S_B$. Each step was assumed to be described by a temperature-dependent Arrhenius rate constant, with two parameters, $E_a$ (the activation energy) and A (the pre-factor). A genetic algorithm was used to fit the above model to the experimental data, and values for the four activation energies and Arrhenius prefactors were obtained.

# Fabrication and Characterization of a Nanocomposite Field-Effect Transistor

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In order to meet society's rising demand for consumer electronics, the development of innovative, inexpensive electronic materials is of paramount importance. Traditional electronic technologies have focused primarily on inorganic semiconductor materials that offer impressive circuit speeds at the expense of their high manufacturing cost and fragility. Organic semiconductors have been the subject of intensive research over the past few decades, as these materials may offer an exciting alternative to inorganics with the attractive attributes of solution processability, greater flexibility, and lower cost. While conduction in inorganic materials is generally much faster than in organics, not all electronic applications require very high switching speeds; active matrix liquid crystal displays, electronic paper, radio frequency identification tags and smart cards are just a few of the many potential applications of organic semiconductors. We propose to combine the easier processability and desirable mechanical properties of organic semiconductors with the superior conduction inherent to inorganics, yielding an entirely new device: the nanocomposite field-effect transistor (NCFET). We anticipate that high aspect ratio inorganic nanostructures will enhance the one-dimensional nature of the current flow through a NCFET, provided that they are aligned parallel to the direction of carrier motion from the source electrode to the drain electrode.


THEORY AND MODELLING

# Modeling Nanoparticle Formation, Growth, and Transport in a Silane Plasma

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Nanoparticles appear in the gas phase of silane plasmas over a range of reactor conditions used during plasma enhanced chemical vapor deposition of silicon thin films. Particles transported to the film may be either harmful or beneficial to film quality, depending on properties such as particle size and number distribution. Numerical modeling of the plasma-particle system provides insight into the processes occurring in the system and aids in parameter optimization for processing a thin film of desired composition.

The spatial and temporal evolution of a RF capacitive silane plasma is simulated in an infinite parallel plate reactor (1-D). An extensive chemistry mechanism describes particle nucleation and includes neutral and negatively charged silicon hydrides, as well as cations, excited species, and carrier gas molecules. Further analysis of the chemistry mechanism eliminates unimportant species, improving the computational efficiency of the model. Coagulation and surface growth are simulated, and the particle size distribution is discretized in a sectional model. Particle charging is also considered, and plasma properties are coupled with both the chemistry and particle modules. Continuum equations are applied to solve the spatial variation in species concentration and include particle transport mechanisms such as gas drag, thermophoresis, diffusion, and electrostatic effects.

# Modeling Chemical Nucleation of Nanoparticles in Fluorocarbon Plasma Systems

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Fluorocarbon plasmas are commonly used for integrated circuit fabrication yet little is known about nucleation and growth of particles in these plasma systems. A zero-dimensional model will predict gas-phase nucleation in these systems accounting for nuclei formation via chemical reactions and particle growth by coagulation and surface reactions. The overall model consists of four interconnected subroutines: base chemistry, plasma model, nucleation, and aerosol model. The base chemistry model specifies the important species and corresponding thermochemical properties; the plasma model determines initial ion and electron densities. Clustering mechanisms are modeled using the nucleation subroutine while surface reactions and coagulation are simulated in the aerosol model.

# Modelling of the Interaction between Fluid Flow and Nanoparticle Dynamics in a Wall Heated Aerosol Reactor

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Wall heated aerosol reactors are well-known devices of generating nanoparticles. Their formation and growth is mainly determined by the thermal decomposition of the precursor and by subsequent processes like surface growth, coagulation, and sintering. Here, the thermal decomposition and particle coagulation are taken into account. As the particles are dispersed in a flow field also particle convection and particle diffusion must be considered. In the present case, four typical aerosol models based on a monodisperse, a moment, a sectional, and a discrete-sectional model approach are considered. The particle distribution in the reactor is investigated and the differences of the four models are emphasised.

The models are compared with respect to the particle size and the particle size distribution. The results vary with the detail of the selected aerosol model and the local position in the reactor. Good agreement is found far away from the reaction zone. In the reaction zone the simple monodisperse and moment model result in larger deviations to the more precisely sectional and discrete-sectional model.

# Numerical Simulation of Particle Transport and Deposition through Filters

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A project in the frame of the SFB 445 is ongoing to simulate the transport and deposition of discrete nano particles.
The gas phase and the electric potential are modeled as continuum by the Navier Stokes equations and the Laplace equation respectively. The governing equations are solved with efficient lattice Boltzmann methods which enable easy treatement of complex boundaries.
The trajectories of the particles arecalculated by integration of the Langevin equation under consideration of drag force, electric force, Brownian motion and van der Waals forces. The discrete consideration of the particles enables the detailed description of the structure and the growth of the deposited layer at the filter surface.
The interaction between the deposited particles and the fluid flow or the electric potential is taken into account by recomputation of these fields after a fixed amount of deposited particles.
Different effects on the particle transport and the deposition are shown by several simulation results.

# Molecular Dynamics Simulations of Dipolar Magnetic Nano Particles

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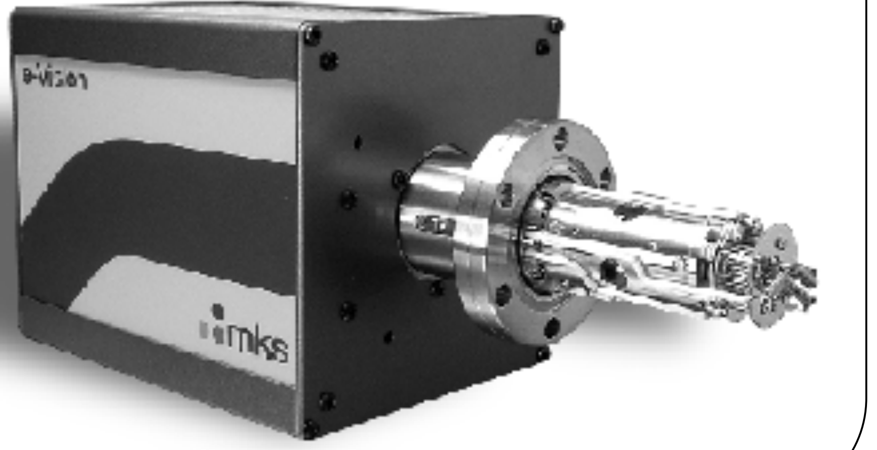
Fe and Ni nano particles grown in the gas phase are known to arrange in chains under certain circumstances. These chains are believed to be stabilized through the long ranged dipolar interaction between the ferromagnetic (or super-paramagnetic) particles.

We investigate the dynamics and thermodynamics of dipolar magnetic nano particles using molecular dynamics simulations. This is done by putting N spherical nano particles in a box with fixed boundaries. We assume that the particles are magnetized along an internal easy axis. Additionally to a hard spheres interaction, the particles interact through the long ranged and anisotropic dipole interaction.

At sufficiently high temperatures $T \gg T_K$, the particles are in the gaseous phase. Approaching the *chain building temperature* $T \to T_K$ from above, the correlations between the particles grow and they start to build chains, which may even clos up to rings for $T \ll T_K$. This transition is accompanied by a peak in the specific heat.

We analyze the influence of the particles magnetic moment, particle size, and temperature on the state of the system.



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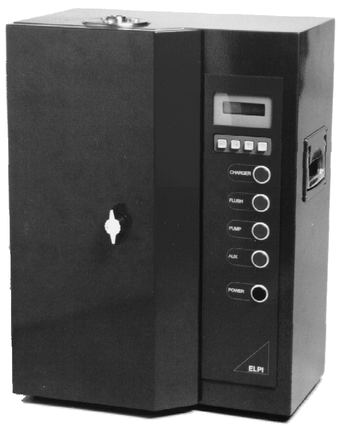
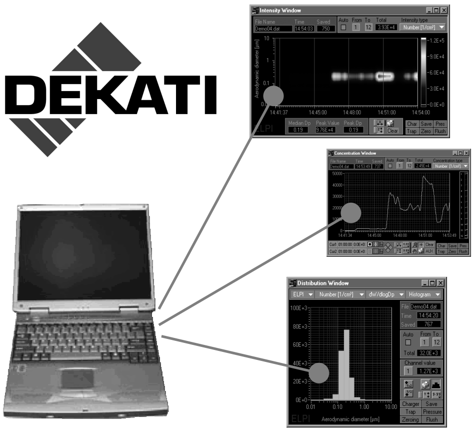
# ELPI
# Electrical Low Pressure Impactor





## Real-Time Particle Size Distribution and Concentration Measurement System

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