Summary

Magnetic nanoparticles are of great interest for researchers from a wide range of disciplines, e.g. catalysis, magnetic data storage, groundwater remediation, and biomedicine. Iron, with high saturation magnetization and abundant reserves, is among the most attractive candidates. The magnetic properties of iron nanoparticles are dramatically different from the bulk matter. The magnetic moment per atom, in near surface regions, drastically changes with the distance from the surface due to discreteness resulting from reduced coordination at the surface, subsequently leading to an improved moment. The downscaling induced property modifications are referred to as finite-size effects that are mainly embodied at two critical points: the single-domain limit and the superparamagnetic limit. Large iron particles adopt a multi-domain structure where regions of uniform magnetization are separated by domain walls.

When the size is reduced below a critical volume, i.e. the single-domain limit typically in the range of several nanometers, it costs more energy to create a domain wall than to support the external magnetostatic energy of the single domain state, and a single-domain structure emerges. Further decrease of the particle size (volume) results in reduction of the anisotropy energy, and the nanoparticles shift to the superparamagnetic state. This is happening when the barrier energy is at a disadvantage in the competition with thermal energy, with the particle behaving as giant magnetic pseudo atoms with huge overall magnetic moment and "collective spin". In this point of view, size control is one of the most crucial and effective means to manipulate the nanoparticle magnetic properties.

Therefore we studied in chapter 3 the controlled synthesis of iron nanoparticles by inert gas condensation. The nanoparticles are constituted by a single crystal iron core, as extensive analysis indicate, and a polycrystalline shell (Fe_3O_4 and/or γ-Fe_2O_3). Particle sizes strongly depend on the gas environment in the cluster source, where the use of helium results in the decrease of size owing to helium's high thermal conductivity. We observed, meanwhile, different nanoparticle shapes including regular and truncated cubic particles, as well as regular and truncated rhombic dodecahedra, which were formed depending on the ratio of the growth rate along the <100> and the <110>direction. The distinct particle shapes can lead to different crystallographic surfaces that enclose the particle with diverse atom densities, electronic structures and bondings, contributing to different physical and chemical properties. Moreover, it was found that iron nanoparticles revealed different morphologies depending on the size.

Cubic shapes are predominant among nanoparticles with a size ~15-24 nm. For particles larger than ~24 nm, the most stable structure is rhombic dodecahedron. Nevertheless, truncated rhombic dodecahedra and cubic shapes with no truncation could also be observed. The facets of iron nanoparticles without truncation exhibit a characteristic inward relaxation indicating that besides thermodynamics also kinetics play a crucial role during particle growth.

Furthermore, in order to obtain a reduced particle size, protective backing plates are positioned in between the iron target and the magnetron, functioning as spacers to modify the magnetic field configuration above the target surface. The decrease of the particle size can be attributed to a reduced effective magnetic field strength combined with a weakened magnetic field confinement. The deposited iron particles with sizes smaller than ~15 nm possess different morphologies from the faceted shapes adopted by larger particles, i.e. they show a close-to-spherical polyhedral shape with a core shell structure for particles sized ~8-15 nm, and a fully oxidized uniform spherical structure with a void in the center for particles sized smaller than ~8 nm.

Finally, imaging of individual spatially localized nanoscale magnets is important, not only for the fundamental physics of nanomagnetism, but also for applications. Therefore, magnetic force microscopy was employed to study the magnetic property and structure of individual iron nanoparticles with sizes ~50-70 nm. The reversal of the magnetic contrasts collected with opposite tip polarities confirms the ferromagnetic state of the studied nanoparticles, and the observed bright and dark contrasts agree with a single-domain structure. Moreover, the magnetization orientation was theoretically simulated with both the particle and the magnetic tip modeled as point dipoles. The non-rectilinear trajectory of the scanning tip, originating from the sharp topographical features of the NPs combined with the tip-apex shape, results in additional side contrasts observed in the phase images. The determination of the remnant magnetization orientation of nanoparticles is beneficial in a multitude of applications where the magnetization of isolated magnetic NPs on surfaces is required.