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ORIGINAL ARTICLE

# Self-organization of nickel nanoparticles dispersed in acetone: From separate nanoparticles to three-dimensional superstructures

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**Abstract** Sonochemical synthesis of monodisperse nickel nanoparticles (Ni-NPs) by reduction of Ni acetylacetonate in the presence of polyvinylpyrrolidone stabilizer is reported. The Ni-NPs size is readily controlled to 5 nanometer diameter with a standard deviation of less than 5%. The as-prepared Ni-NPs sample was dispersed in acetone, for 4 weeks. For structural analysis was not applied to a magnetic field or heat treatment as key methods to direct the assembly. The transition from separate Ni-NPs into self-organization of three dimensions (3D) superstructures was studied by electron microscopy. Experimental analysis suggests that the translation and rotation movement of the Ni-NPs are governed by magnetic frustration which promotes the formation of different geometric arrangements in two dimensions (2D). The formation of 3D superstructures is confirmed from scanning electron microscopy revealing a layered domain that consists of staking of several

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monolayers having multiple well-defined supercrystalline domains, enabling their use for optical, electronic and sensor applications.



## 1. Introduction

The organization and self-assembly of nanostructured materials with controlled size and composition are fundamental with a technological interest over recent years. These kinds of materials provide the critical building blocks for nanoscience and nanotechnology exhibiting new and enhanced properties compared to bulk materials. The effort to understand the physical properties of ever smaller structures has been paralleled by attempts to exploit their beneficial properties. Indeed, in the field of magnetic particles, physical parameters defined as constants in descriptions of bulk materials properties, become size dependent [1]. In other words, changing the size and shape of nanoscale materials produces new materials. Furthermore, the self-assembly of magnetic nanoparticles can lead to superstructures with important collective properties such as data store device [2], spin-dependent electron transport [3], photonic crystals [4], tandem catalysis [5], and a rich variety of novel phenomena derived from their collective interactions.

In this context, the self-assembly of Ni-NPs into three dimensional (3D) superstructures arouse intensive interests from the perspective of both fundamental research and application purpose. Reports on the preparation of Ni nanoparticles self-assembly are not common compared to numerous reports on the preparation of 3D superstructures such as CdSe [6,7], Au [8–10], Ag [11–13], and Co [14,15]. Also, the development of synthetic routes has been employed to synthesize Ni nanoparticles in both organic and aqueous media [16–18] and surfactants are generally used to synthesize monodispersed Ni-NPs [19–22]. However, very limited information is available on the transition from separate Ni-NPs to self-organization in aggregates at a microscopic scale. Therefore, the present study focuses on the synthesis of Ni-NPs with tunable sizes using a simple sonochemistry synthesis in the presence of polyvinylpyrrolidone (PVP) stabilizer. The as-prepared sample was dispersed in acetone and the interplay between crystallographic ordering and its dynamics were discussed herein.

## 2. Experimental

### 2.1. Reagents and materials

All chemicals used in this experiment were of reagent grade. Ni $(OCOCH_3)_2 \cdot 4H_2O$ ($\geqslant$98%, Aldrich) was used as nickel source, acetone ($\geqslant$99.9%, Aldrich), isopropyl alcohol ($\geqslant$99.7%, Aldrich) and Poly(vinyl pyrrolidone, PVP, $M_w$ = 40,000) were used as received without further purification. The water used throughout this work was deionized water.

### 2.2. Synthesis of nickel nanoparticles

Nickel nanoparticles were synthesized as follows: 1 g of Ni $(OCOCH_3)_2 \cdot 4H_2O$ was dissolved in a mixture of 10 mL of acetone and 10 mL of Isopropyl alcohol. The reaction mixture was exposed to ultrasound irradiation (20 Khz) for 20 min. At this time, 0.05 g of PVP was added to the mix and the ultrasonic treatment extended until 50 min [23] was completed. The reaction formed a black solid, which was separated by centrifugation and washed several times with acetone and isopropyl alcohol and dried at room temperature. The as-prepared sample was redispersed in 15 mL of acetone. The suspension of Ni-NPs was preserved inside a vertically positioned glass vial for a maximum time of 4 weeks. After this time, five samples were obtained and they are identified by its week number (as-prepared, W1, W2, W3 and W4) along this paper. It is important to remark that no heat treatment was implemented after the sonochemical synthesis of Ni-NPs in order to preserve the crystalline structure and the crystal size of the as-prepared sample.

### 2.3. Characterization

X-ray diffraction (XRD) patterns were carried out with a Bruker X-ray diffractometer D8 Focus ($\lambda$ = 1.5406 Å) with 35 kV and 25 mA. High resolution transmission electron microscopy (HRTEM) and a scanning transmission electron microscope (STEM) operated in the mode in which the scattered electrons are collected by means of a high-angle annular dark-field (HAADF) detector to provide the critical experimental input needed to determine the full-structure evolution of the assembled Ni-NPs. Therefore, the morphology and structure were characterized on a Tecnai F30 ($Cs$ = 1.2 nm) operated at an accelerating voltage at 300 kV having a point-to-point resolution of 0.20 nm and lattice resolution of 0.14 nm. Samples for HRTEM and STEM crystallographic analyses were prepared by evaporation of one drop of Ni-NPs solution on carbon-coated copper grids (300 mesh) and then dried under air. Morphological evolution analysis was conducted on a scanning electron microscopy (SEM, FEI-Nova 200 Nanolab) operated at 10 kV.

## 3. Results and discussion

The corresponding XRD spectrum for the as-prepared sample (Fig. 1) clearly shows the presence of a single face-centered cubic (FCC) crystal phase of Ni NPs with only three reflections, ($2\theta$ = 44.6°, 51.84° and 76.48°), corresponding to Miller indices (1 1 1), (2 0 0) and (2 2 0) respectively (JCPDS 04-0850). No peaks of nickel oxide were detected within the XRD analysis limit, indicating that pure FCC nickel was obtained under such experimental conditions. The nanometric crystallite size can easily be detected by the broadening of the (1 1 1) reflection, due to that the XRD reflections are usually inversely correlated with the crystallite diameter of the particles. The size of coherently diffracting domains was calculated by considering the size broadening contribution only and to determine the grain size based on the (1 1 1) reflection the Debye-Scherrer