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Numerical simulation of magnetizing processes of nickel nanowires and nanoballs grown in superfluid helium

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Abstract. The present work is dedicated to numerical modeling of magnetization processes of Ni nanoparticles ensembles acquired by pulsed laser ablation of metallic target in He II. The characteristic feature of the systems under study is their heterogeneous composition. One fraction of particles is represented by thin nanowires (with diameters of 4 nm strictly) meanwhile the other corresponds to a ball-shaped particles that can obtain diameters from 8 to 500 nm. The model was verified by comparing its calculation results with SQUID-magnetometry data.

1. Introduction

The magnetic properties of nanoobjects are a very interesting object for research, both from a fundamental and from an applied point of view. On the fundamental side, it is important to understand how the parameters that determine the hysteresis of the magnetization of ferromagnetic particles depend on their diameter. From engineering point of view, of great interest is the problem of creating materials that can provide a high recording density of information, which is associated with progress in miniaturization of computer and memory technologies [1, 2].

The universal method of nanowires synthesizing by means of pulsed laser ablation (PLA) in superfluid helium (He II) implemented by our group allows to obtain quasi-1D objects made from almost any material including ferromagnetic ones [3, 4]. Usually PLA produces spherical nanoparticles with lognormal size distributions, but in He II due to its unique properties the ablation products are condensing also as nanowires consisted of thinnest nanowires. A diameter of the nanowire is determined only by thermophysical parameters of ablated material and is distributed with almost zero variance. Our experiments reveal the considerable variety in magnetic hysteresis properties of such systems. The aim of current work is to explain this phenomenon.

2. Experimental

The scheme of setup used for synthesizing of Ni nanowires is shown in figure 1. We used the pulsed solid-state Nd:LSB laser with wavelength $\lambda = 1.064 \mu\text{m}$, pulse energy $E = 0.1 \text{ mJ}$, pulse duration 0.4 ns, and repetition rate of 4000 Hz. Target made from nickel foil was placed inside of helium cryostat and was merged in the liquid He II during the ablation. Laser beam focused by a lens was introducing into



the cryostat through the sapphire window. The ablation products after condensation in He II volume sedimented on flat single-crystal Si substrates (1x1x4 mm) as well as on standard perforated 3 mm TEM meshed grids both located below the target. The ablation duration which affects on the concentration of Ni products on the substrates was 15 to 60 minutes for different experiments. For a day after experiment the cryostat warmed up following which the specimens on Si substrates were transported to SQUID-magnetometer MPMX 5XL (Quantum Design) and SEM microscope, and TEM-meshes transported to transmission electron microscope JEM-2100 (JEOL). The magnetic moment measurements were carried out at room temperature.

The numeric simulation of magnetic properties was realized in MATLAB.

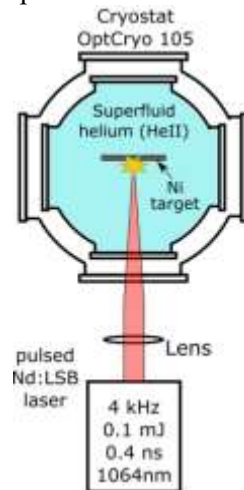


Figure 1. Scheme of experimental setup (cross-section top view)

3. Experimental results

The TEM images of nanosystems under study is shown in figure 2. According to the microscopy data the nanosystems are represented by webs of nanowires (Ni wire diameter ~ 4 nm) with spherical particles inclusions having diameters in range from 5 to ~ 250 nm with lognormal distribution of sizes (figure 3) typical for laser ablation methods. The spherical particles are single-crystal whereas the nanowires are polycrystalline with grain sizes of ~ 1 nm.

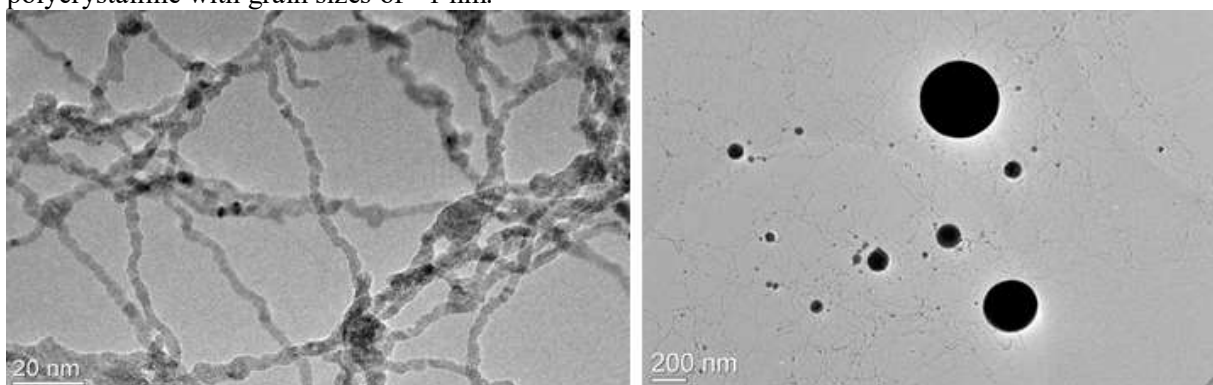


Figure 2. TEM images of Ni nanosystems obtained by laser ablation method in He II.

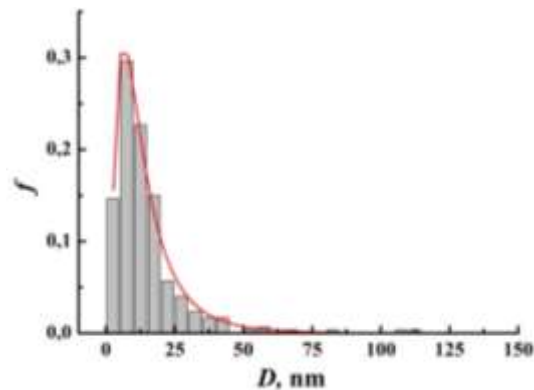


Figure 3. Normalized empirical distribution f of diameters D of Ni spherical particles acquired during the analysis of the SEM data (sample group of 300 particles) and its lognormal approximation.

The magnetometry of four specimens arranged in ascending order (from a to d) according to the total magnetic moment are represented in figure 4. It is seen that one of them differs from others qualitatively by high value of coercivity H_c and low value of saturation magnetic moment M_s which entails almost square hysteresis loop (figure 4a). Other specimens demonstrate s-shaped hysteresis with low H_c and high M_s (figure 4b, c, d). It seems important to emphasize that the case of square loop corresponds to the minimal average concentration of nanoparticles on the Si substrate according to the M_s value.

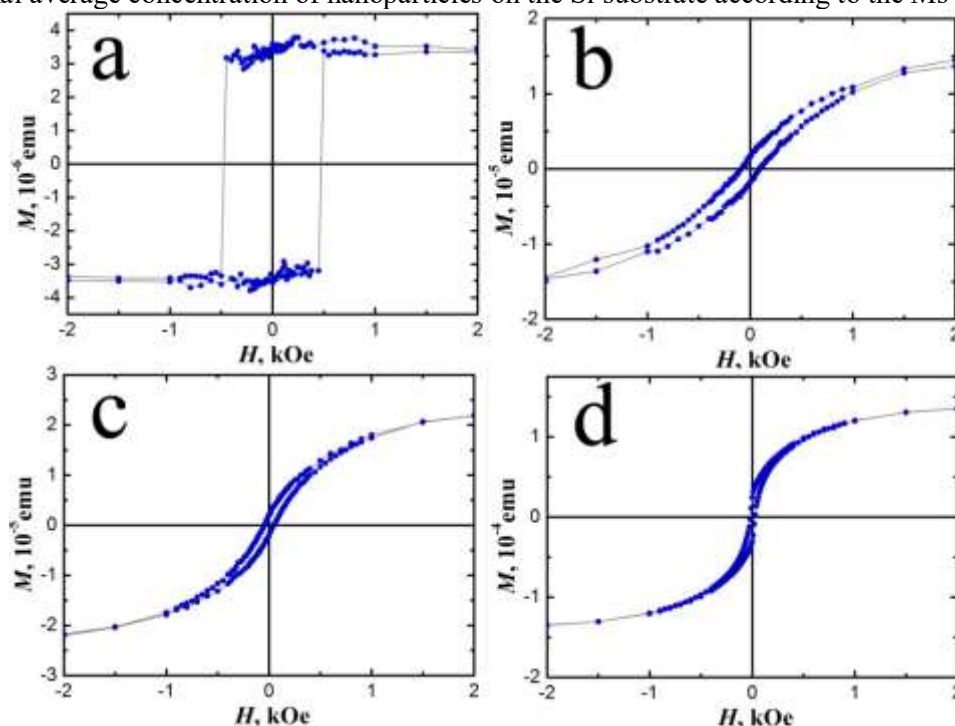


Figure 4. Experimental magnetization curves $M(H)$ of four specimens (a - d) of Ni nanosystems on Si substrates measured at 300 K in-plane (diamagnetic contribution of Si is subtracted).

4. Calculation method

The basic assumptions adopted in order to simulate the magnetic behavior of systems under study are:

- the complex nanosystem can be represented as an ensemble consisted of individual particles of two types: nanowires and spherical nanoparticles;

- interaction between two any particles from an ensemble is absent. This assumption implies possibility of modeling a magnetic hysteresis of any individual particle separately which allows to carry out summation in any convenient order and over a set of particles of interest to acquire the resultant loop;
- shape and magnitude of hysteresis loop of individual particle can be described mathematically using some expression with small number of parameters.

The assumptions allow to separate the contributions of nanowires and spherical particles in total magnetic moment as well as approaches to the modeling of these contributions.

4.1. Nanowires hysteresis modelling

For every particle in this case the hysteresis was assumed to have square shape [5] with half-width H_c and half-height M_s . The orientation of every nanowire relative to the external field H was assumed to be constant random number lying within some adjustable range of disorientation angles. The magnetic moment of every nanowire was assumed not to leave the direction of anisotropy axis (which matches nanowire axis because of shape anisotropy) jumping to the opposite direction upon the projection of magnetizing field H reaching certain value. The contribution of every nanowire to the total magnetic moment was calculated as the projection of its individual magnetic moment on the direction of external field. For simplicity it was also assumed that magnetic moment of every nanowire depends only on the number of Ni atoms of which it consisted of, the moment of atom being equal to 1, and the average length of nanowire (170 nm) and diameter on each nanowire (4 nm) being known from the SEM microscopy data. Following these assumptions, the individual loops for every particle were calculated, summated over all nanowires (10^4), normalized by dividing on the total calculated M_s and thus we obtained the resultant loop.

4.2. Spherical particles hysteresis modelling

The loop shape $M(H)$ for individual spherical particle was described by three-parameter sigmoid functions:

$$M(H) = M_s - \frac{2M_s}{1 + \exp\left(\frac{H \pm H_c}{h}\right)} \quad (1)$$

Here «+» and «-» signs correspond to descending and ascending branches of hysteresis loop, and parameters M_s , H_c , h are introduced and interpreted as saturation magnetic moment, coercivity and slope coefficient. The parameters were calculated regarding to the size of the particle in the following way.

Saturation magnetic moment M_s of particle was assumed (as in the case of nanowires) to depend only on the number of Ni atoms in corresponding particle while magnetic moment of atom was chosen as unit. Coercivity H_c for the particle of known radius was calculated according to the theoretical curve (figure 5), which was referred to our case by two additional assumptions: superparamagnetic limit is 1 nm and single-domain radius $R_0 = 30$ nm under coercivity 200 Oe.

Parameter h corresponding to the common slope of the loop is related to the magnetic susceptibility of a spherical particle. Although the search of publications on that subject has left to nothing, we have empirically found the following expression to be satisfying:

$$h(R) = \alpha + \frac{\beta}{R^2} \quad (2)$$

Coefficients α and β were chosen manually to provide optimal matching of calculation results with experiment.

After finding of the appropriate (for every particle of known size) set of parameters (M_s , H_c , h) the $M(H)$ curve were calculated for them to be next scaled with regards of number of particles of that size, which was chosen according to the lognormal distribution (3)(figure 3):

$$N(R) = \frac{1}{\sqrt{2\pi}wR} e^{-\frac{\left(\ln\left(\frac{R}{x}\right)\right)^2}{2w^2}}, \quad (3)$$

where w , x are the numeric parameters of distribution, varying which one can model different possible empirical size distributions of spherical particles. All obtained loops were then summed and normalized to one by the value of total magnetic moment.

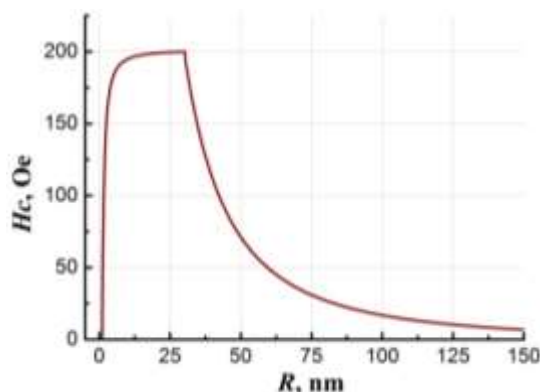


Figure 5. Size dependence of coercivity of individual spherical particle [6].

5. Results and discussion

Square shape and high H_c value for the hysteresis shown in Figure 4a let one to assume that corresponding sample consists predominantly of the particles with strong magnetic anisotropy which are in our case nanowires. That's why we have assumed that the whole ensemble consists of nanowires of the same (and equal to average) length for that case, allowing only that the degree of disorientation in ensemble may be different.

As it is seen from Figure 6a, the last is affecting significantly only on the value of saturation magnetic moment, because both calculated curves match experimental ones more or less satisfyingly. However, they have their advantages and disadvantages needed to be discussed. The curve which corresponds to higher disorientation range explains nonzero loop thickness in the area of high fields, but in the area of low fields it doesn't simulate the experimental data well. At the same time the curves which correspond the partial disorientation of particles simulate the experimental data more and more accurately with decreasing of disorientation range, but they do not predict the behavior of the loop at high fields and also this case requires presence of some force, which would orient the wires on the substrate. Of course, the external field H can play that role.

In turn to explain s-shaped hysteresis loops in figure 4(b-d) the predominant impact of spherical particles was assumed in corresponding samples. In concordance with this for explanation of experimental data the ensembles of spherical particles with radii from 2 to 250 nm were considered. The previously found from approximation (figure 3) of empirical distribution of spherical particles on the substrate values x , w were chosen as initial estimate for the size distribution. For value w determines the proportion of large and small particles in ensemble, the next step to take was to adjust it to provide best matching of H_c for each loop (figure 4b, c, d) considering the experimental accurateness of determining H_c . In addition, the values $\alpha = 5$; $\beta = 700000$ were manually found for expression (2).

The results of the approach are shown in Figure 6b, c, d. It is seen that H_c behavior at low and high fields for calculated and experimental curves are in good agreement. However, the areas confined inside the calculated curves are overestimated as compared to the experimental ones in all cases. The divergence can possibly be explained by non-considered interaction between particles which can affect the total energy required for remagnetization of the system (known to be related to the element of the loop area $\delta M \cdot \delta H$).

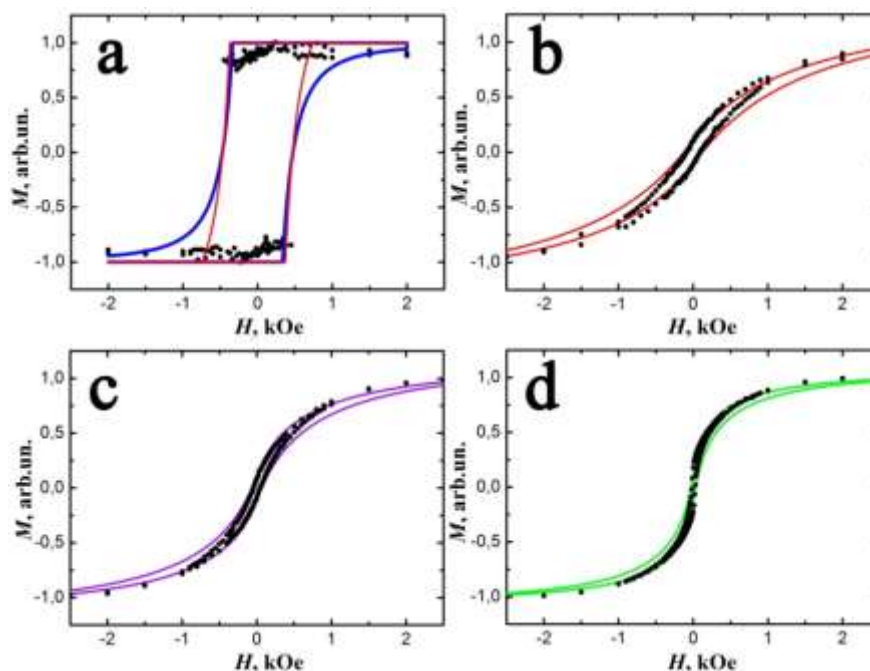


Figure 6. Comparison of experimental (dots) and calculated (lines) data. The samples order is correspondent to one of the Figure 4. The heavy line in (a) corresponds to complete disorientation of nanowires, thin line corresponds to partial disorientation.

6. Conclusion

The magnetic properties of Ni nanosystems grown by means PLA in He II are appreciably dependent of the proportion of spherical and filamentary particles in particular sample. Present data suggest that the proportion can be affected by a number of random factors. Nevertheless, magnetic properties of discussed systems can be successfully reproduced by applying a simple model suggested. As follows from the model, the square hysteresis is in the good correspondence with the ensemble of highly anisotropic nanowires, whereas s-shaped hysteresis is the consequence of spherical particles predominance. It's also important to note that the rareness of experimental square hysteresis cases is then finding its natural excuse, because the presence of even several large ($R \sim 250$ nm) spherical particles is enough to overlap the signal from all the thin nanowires on the substrate. The model also opens the principal opportunity to set and solve the inverse problem that is to find out the constitution of the nanosystem knowing the parameters of its hysteresis and some information on the types of particles present.

Acknowledgements

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