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Magnetic-particles-composed wire structures produced by pulsed laser deposition in a magnetic field

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Abstract. We demonstrate the possibility to fabricate wire structures composed by arranged magnetic particles using pulsed laser deposition (PLD) in the presence of a magnetic field. Ablation of Ni and Co targets was performed in air by nanosecond laser pulses delivered by a Nd:YAG laser system oscillating at 355 nm. Due to the high density of the ambient, particles and clusters were formed by condensation in the plasma plume close to the target. The strong deceleration of the ablated material under these conditions further benefited the efficiency of applying a magnetic field to the plume. We also studied the effect of the target-to-substrate distance and the ambient pressure on the morphology of the deposited structures.

1. Introduction
Magnetic micro- and nanostructures continue to sustain the interest of researchers due to their potential use in areas such as high-density data storage, biomedical applications, drug delivery, magnetic separation and sensors [1–7]. The properties of these structures differ from those of their bulk counterparts because of their high surface-to-volume ratios. Moreover, such structures could also exhibit novel properties influenced by their arrangement and the ability to form complex systems. When an external magnetic field is applied to an ensemble of micro- or nanoparticle, alignment of the particles’ magnetic moments can take place and, as they interact, mask-free assemblies of 1D, 2D or 3D systems can be produced [8]. The possibility of additional control of the system orientation on a macroscale by the external magnetic field can result in these structures acquiring unique tunable directional-dependent electric, magnetic and optical properties [9].

The magnetic structures’ unique characteristics led to the development of a wide range of fabrication methods, including microemulsions, sol-gel synthesis, sonochemical reactions, hydrothermal reactions, hydrolysis and thermolysis of precursors, flow injection synthesis, and electrospray synthesis [10–16]. However, these chemically-based techniques usually involve several steps and often require toxic reagents. Even in the well-developed chemical, physical and combined methods, the cost-efficient, easy-to-implement and time-saving fabrication of 2D and 3D magnetic micro- or nanostructures is still a challenge.

As a flexible method, the pulsed laser deposition (PLD) has proven its capability to produce complex structures of a wide variety of materials. The technology includes ablation of a target and deposition of the ejected material on a substrate. The morphology of the deposited structure can be

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controlled via different experimental parameters: laser processing characteristics, deposition geometry, substrate temperature and environment parameters. Besides the conventional parameters, the application of an external magnetic field to the plasma plume has also been established as being an important parameter in the PLD technique. It was demonstrated that the absence of a magnetic field during laser ablation of gold in water leads to the formation of nanowires with a length of \( \sim 100 \) nm, while single nanoparticles and shorter nanowires (\( \sim 10-20 \) nm) are formed when a magnetic field is applied [17]. Recently, ordered magnetic nanostructures were produced by femtosecond laser ablation of steel in air and in the presence of an external magnetic field [18]. The specific dynamics of the ejected material under these conditions leads to the formation of nanoparticle wires with lengths of tens of micrometers on the substrate. Despite the great possibilities of the method, the need for a femtosecond laser system limits the flexibility of the method, which hampers its efficient application in the industry. Furthermore, fabricating magnetic structures with a desired orientation by nanosecond laser ablation in the presence of an external magnetic field still presents considerable challenges.

In this paper, we present results on the nanosecond laser ablation of Co and Ni when a magnetic field is applied. We found that the deposition process produces wire structures composed of arranged particles. We studied and discussed the effect of varying the target-to-substrate distance on the morphology of the formed structures, and how the ambient pressure affected the arrangement of the deposited material.

2. Experimental
The fabrication of wire structures was implemented by applying the PLD process in the presence of external magnetic field. The targets of ferromagnetic metal (Co—99.99% or Ni—99.99%) were mounted on a rotating holder and ablated by nanosecond laser pulses delivered by a Nd:YAG (Lotis LS-2147) laser system operating at the wavelength of 355 nm. The pulse duration was 15 ns at a repetition rate of 10 Hz. The angle between the incident radiation and the target surface was about 30°. The deposition process was performed in a standard on-axis geometry (figure 1) with laser fluences of 20 J/cm\(^2\) or 45 J/cm\(^2\) and deposition times of 2 min and 5 min. The ablated material was deposited on a quartz substrate fixed at 5 mm or 3 mm from the target. A permanent magnet producing a magnetic field of \( B = 0.2 \) T was attached to the back side of the substrate. The depositions were conducted in air at atmospheric pressure or in He at pressure of 500 Torr. The composition of the material deposited on the substrate was analyzed by EDX (Quantax 200 Bruker). The morphology of the structures produced was studied by SEM (LYRA Tescan).

3. Results and discussion
Figure 2 shows SEM images of the material deposited after ablation of a Co target in open air, without application of a magnetic field (a) and in the presence of a magnetic field (b). The ablation was performed at a laser fluence of 20 J/cm\(^2\) with a deposition time of 5 min in both cases. The distance between the target and substrate was 5 mm. As seen, deposition without an external magnetic field applied leads to the formation of a structure consisting of randomly distributed nanoparticle aggregates and free nanoparticles with diameters up to 100 nm. The origin of these nanostructures is related to the high density of the ambient atmosphere. In the case of deposition in air at atmospheric pressure, the nanoparticles and nanoparticle aggregates are formed by condensation close to the target [19]. When the magnetic field is applied during the deposition process, distinct wire structures with lengths of several tens of microns are formed on the substrate (figure 2b). The orientation of the wires is predominantly parallel to the magnetic field applied. It should be mentioned that the spatial
confinement of the plasma plume and the drop in the particles’ velocity further benefit the efficiency of application of the magnetic field on the flying material. A high-magnification SEM image of this sample reveals the presence of free nanoparticles (inset in figure 2b). Similar to the case without a magnetic field, the nanoparticle substructure has a random distribution over the substrate. The lack of arrangement of these particles is related to their small size and the high temperatures they acquire in the plasma plume.

**Figure 2.** SEM images of Co structures produced by PLD in open air, without application of an external magnetic field (a) and in the presence of an external magnetic field (b). The samples are produced at a laser fluence of 20 J/cm², deposition time of 5 min and target-to-substrate distance of 5 mm. The inset in (b) is a high-magnification image of the sample.

The composition of the structures produced by PLD of Co in open air was revealed by EDX analysis. Figure 3 is an EDX spectrum taken from the structure shown on figure 2b. It should be noted that besides Co, a peak corresponding to oxygen is also observed in the EDX spectrum. The presence of oxygen in the sample is related to oxidation of the top layer of the Co wires due to the high concentration of surface defects.

**Figure 3.** EDX spectrum (right) of a wire structure produced by PLD of Co in open air and external magnetic field applied. A SEM image of the structure (left) is also shown. The laser fluence is 20 J/cm², the deposition time is 5 min, and the target to substrate distance is 5 mm.

Further, we explored the influence of the process parameters on the morphology of the structures deposited in the presence of an external magnetic field. Figure 4 shows SEM images of samples
deposited at different target-to-substrate distances – 5 mm (a) and 3 mm (b). The depositions were performed in open air at a fluence of 45 J/cm² and a deposition time of 5 min. Clearly separated wires oriented along the external magnetic field are observed in both structures. However, the Co wires morphologies in the two samples differ significantly. When the target to substrate distance is 5 mm, the deposition process results in elongated features with a length ranging from 25 to 45 µm and a width of 4-10 µm. In the case of deposition at 3 mm, fine wires with lengths in the range from 8 to 35 µm and a width of 2-3 µm are formed. The formation of thicker structures in the sample deposited at 5 mm could be attributed to the higher degree of aggregation of the ablated material due to the longer impact of the magnetic field on the plasma plume compared to the deposition at 3 mm.

**Figure 4.** SEM images of Co wire structures produced by PLD in open air and an external magnetic field applied. The target to substrate distance is (a) 5 mm and (b) 3 mm. The laser fluence is 45 J/cm² and the deposition time is 5 min. The inset in (b) shows a high-magnification image of the structure.

In order to examine the capabilities of the method utilized, we also performed deposition of Ni in a magnetic field. Figure 5 presents SEM images of the material after the ablation of a Ni target in air at atmospheric pressure (a) and in He at 500 Torr (b). The experiments were carried out under the same conditions as the structure shown in figure 4a. A shorter ablation time was used for the sample.

**Figure 5.** SEM images of Ni structures produced in the presence of an external magnetic field by PLD (a) in air at atmospheric pressure for a deposition time of 5 min, and (b) in He at pressure of 500 Torr for a deposition time of 2 min. The laser fluence is 45 J/cm², and the target to substrate distance is 5 mm. The insets show high-magnification images of the structures.
produced at 500 Torr due to the more efficient deposition of material compared to the case of ablation at atmospheric pressure. As seen, the application of an external magnetic field during PLD of Ni also affects the morphology of the deposited material. The deposition at atmospheric pressure results in wires composed of particles arranged along the magnetic field, as well as in free particles (figure 5a). The particle wires formed have lengths from 10 to 40 µm and widths of 2-4 µm. Similar elongated features are observed in the case of deposition in He at the lower pressure, but they are significantly less magnetically-oriented and rather randomly distributed (figure 5b). The lower degree of orientation is related to the pressure of the ambient medium. A lower ambient pressure will lead to a higher velocity of the ablated material, which will limit the time of the external magnetic field acting on the plasma plume, so that the efficiency of the orientation process will be reduced.

4. Conclusions

We described a technique for fabrication of 2D wire structures composed of arranged magnetic particles. The method is based on the implementation of the PLD process in the presence of an external magnetic field. The ablation of Co under these conditions leads to the formation of distinct wire structures with lengths of several tens of microns. The wires on the substrate are predominantly oriented along the direction of the magnetic field applied. When no magnetic field is applied during the deposition process, randomly distributed nanoparticle aggregates and free nanoparticles are formed on the substrate. We found that the size of the Co wires strongly depends on the target-to-substrate distance. When this distance is 5 mm, thick elongated structures with lengths in the range from 25 to 45 µm and widths of 4-10 µm are produced. The deposition at a 3-mm distance results in fine wires with lengths ranging from 8 to 35 µm and width of 2-3 µm. The ablation of Ni in a magnetic field also leads to the creation of oriented wire structures. The Ni-wires length ranges from 10 to 40 µm, the widths being 2-4 µm. The technique presented could find applications as an inexpensive and flexible way for fabrication of new materials and structures.

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