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# Significant tunability of thin film functionalities enabled by manipulating magnetic and structural nano-domains

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ABSTRACT

The influence of laser frequency on the structure and physical properties of thin films grown by pulsed laser deposition has been studied. Different types of thin films, hard ferromagnetic FePt $L1_0$ and quasi-single crystal superconducting $YBa_2Cu_3O_7$ (YBCO), have been used for demonstration of the effect. Significant structural modifications have been obtained for the films with similar thicknesses. These modifications are shown to dramatically control their corresponding properties, providing an instrumental ability for tuning the practical characteristics of the films by changing the laser frequency of their deposition. In particular, 20-fold increase of coercive field and modification of demagnetization mechanism are obtained for FePt films by varying the frequency from 1 Hz to 6 Hz. Over a similar frequency range, a strong dependence on the laser frequency is discovered for the YBCO films for the critical current density behavior as a function of the applied magnetic field [$J_c(B_a)$] with the unexpected reversal of $J_c(B_a)$ curves with temperature. The mechanisms of structure modifications and corresponding property variations are proposed.

## 1. Introduction

Simple and reliable methods with the high degree of technological control are required for accurate adjustments of material structures and corresponding properties for the required performance. For physical vapour deposition (PVD) technologies, this control is conventionally achieved by manipulating interdependent parameters, such as temperature, atmosphere, flux of material, resultant film thickness [1–6] and laser frequency [6–10]. In particular, the laser frequency for pulsed-based deposition (PLD) influences the film structure in many different ways, whose mechanisms can be contentious and not easily understood [7–11]. Thus, this interdependency is not trivial, impeding reproducibility and requiring sophisticated control with the help of, for example, reflection high-energy electron diffraction (RHEED) technique.

This work is focused on tuning the structure and properties of thin films through the variation of laser frequency. It turns out that the laser frequency is convenient and powerful tool for applications in thin film technologies.

Two model materials are chosen to study the influence of the laser frequency: $L1_0$ FePt hard ferromagnetic thin films and quasi-single-crystal superconducting $YBa_2Cu_3O_7$ (YBCO) thin films. It should be noted that these model films are completely different: (i) YBCO is a perovskite type metal-oxide, while FePt is a metal with *fct* superlattice; (ii) for the best performance, a FePt film is required to consist of well separated islands of tens to hundreds of nanometers large, whereas YBCO should have a quasi-single-crystal structure completely covering substrate and be as smooth as possible; (iii) FePt films grow via typical Volmer-Weber island growth [9,12], while YBCO is grown by Stranski–Krastanov mechanism [13–15] as the result of the film composition and the crystal lattice mismatch with the substrate ($SrTiO_3$, in our case).

Quasi-single-crystal *c*-axis oriented epitaxial YBCO thin films are known to have the highest critical current density ($J_c$) among high-temperature superconductors at temperature $T = 77$ K [16,17]. It makes YBCO thin films a perfect candidate for various electric power applications [18–20]. In addition, applications of YBCO thin films are associated with microwave electronic devices [21,22] and low signal electronic devices [19,23,24]. The critical current density of these films (like any type-II superconducting material) is governed by pinning of superconducting vortices by crystal defects. The strongest pinning sites consist of columns of a

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non-superconducting material with the diameter close to coherence length, and that are aligned with the direction of applied magnetic field ($B_a$)[25–27]. For high quality YBCO thin films made by physical vapour deposition techniques, the out-of-plane edge dislocations are alike columns, which are formed in situ during growth of the films [27]. The arrays of these dislocations form low-angle domain boundaries [13,15,28–30]. A very large density of these pinning sites is required for high critical current densities. On the other hand, these pinning sites, created by disturbed crystal lattice parameters, possess suppressed superconducting order parameter. The superconducting order parameter defines the capability of the material to form the superconducting charge carriers (Cooper pairs). Hence, an optimal oxygen content and the best possible crystallographic order of YBCO films are necessary to reach high and homogeneous superconducting order parameter (see, for example[31,32]). Various conditions relevant to applications, such as external magnetic fields and temperatures, can strongly affect the order parameter and pinning properties, which in turn can strongly influence the superconducting performance. Thus, fine tuning of deposition conditions is required to reach the best performance in particular conditions (for particular applications), where the deposition frequency can play the key role.

Furthermore, a critical thickness for the optimal YBCO performance exists [5,6,33,34]. In thicker films, the surface becomes rough and their properties degrade, unless a multilayered structure is introduced [35]. In thinner films, the integral capabilities of the films are reduced. The optimal thickness, defined at the maximum of the critical current density ($J_c$) as the function of the film thickness [5,33], is usually obtained at the deposition stage with all YBCO islands coalesced [5,6,34]. In this work, it is shown that the island density and the island growth kinetics can be effectively varied by changing deposition conditions (in particular, laser frequency), thus the optimal thickness can be adjusted at will.

FePt thin films with $L1_0$-*fct* superstructure are considered for various existing and emerging applications, such as perpendicular magnetic recording media (MRM) by Seagate [36], microelectromechanical systems (MEMS) [37], ultra strong permanent magnets [1,2,38], or ferromagnetic-superconducting heterostructures [39]. These applications become possible due to the film ability to form thermally stable islands (or grains) down to a few nanometers [38,40,41] (boosting recording density [42]), their high magnetic anisotropy, very high coercivity (exceeding 10 T) [1] and magnetic energy product, the out-of-plane magnetization, and high corrosion resistance. The strong magnetic properties of the films can also be employed for the compensation of applied magnetic field induced effects [39] in certain architectures combining ferromagnets and superconductors [43,44]. The behavior resulting from the interactions can extend magnetic screening (Meissner effect) [45] and even enhance current carrying ability in superconducting wires [46].

In this work, a unique advantage of pulsed laser deposition (PLD) technique in tuning desirable structures and properties of the FePt and YBCO epitaxial films is demonstrated. To the best of our knowledge, such dramatic structure variations and such a broad range of practically achievable physical properties, being tuned by means of mere laser frequency variation, have not been reported before. Note, the key instrumental feature for varying properties of the films is the ability to manipulate the in-plane structure of the films, while *their thickness is kept constant*. In the case of YBCO thin films, it allows one to adjust critical current density dependence on applied magnetic field ($J_c(B_a)$) for different applications at different operating temperatures. In the case of FePt thin films, it facilitates the adjustment of the coercive field and magnetic domain structure. Our new approach is in contrast to the commonly used method for the modulation of island structure of the FePt films by means of the thickness variation, which is proportional to the size of the islands [1–3]. However, making these films too thin may reduce the read-back signal below the detectable threshold and the magnetic energy product for permanent magnets.

## 2. Experimental details

### 2.1. PLD of YBCO and FePt thin films

Quasi-single-crystal *c*-oriented YBCO thin films were produced on 5 × 5 mm size $SrTiO_3$ (STO) substrates using standard pulsed-laser deposition (PLD) [33,35]. Deposition was performed by KrF excimer laser with the wave-length of 248 nm. The laser fluency was kept at $\sim 3 - -4$ J/cm$^2$, but taking into account optical path losses and laser beam homogenization, the fluency of $\sim 2$ J/cm$^2$ is a more realistic estimation. The frequency of the laser was varied from 1 to 8 Hz.

For the deposition of YBCO films, the PLD chamber was pre-evacuated down to $10^{-6}$ Torr. During the deposition, the substrate temperature and background oxygen pressure were 780 °C and 300 mTorr, respectively. Distance between the substrate and the target was 85 mm. The films where annealed in oxygen atmosphere at 400 °C for one hour after deposition for oxygenation. The thin films measured in this work are of optimal thickness $d_p \simeq 300$ nm thick[5,6,33], measured by Dektak profiler. The critical temperature was $T_c \simeq 90.0 \pm 0.5$ K for films deposited with 5 and 8 Hz frequencies and $T_c \simeq 91.5 \pm 0.5$ K for 1 Hz deposited film, measured by DC magnetic measurements in PPMS at $B_a = 2.5$ mT. FePt thin films with $L1_0$ *fct c*-oriented islands were deposited on the same type of the STO substrates using the same PLD system. The justification for the substrate material choice can be found elsewhere (see Ref. [11] and references therein). The base pressure in the PLD chamber was typically held at $\leq 10^{-8}$ Torr that was reached after additional PLD chamber cleaning and baking routine. The pulsed laser deposition of the films was carried out with the substrate temperature held at 800 °C, which degraded vacuum during deposition down to $\sim 5 \times 10^{-7}$ Torr due to thermal desorption from the heater. The deposition temperature was chosen to ensure that the $L1_0$ structure of the FePt alloy forming at 500 °C [47–50] was complete [2,37,50–53]. The deposition process, facilitating almost equiatomic composition in the FePt epitaxial superstructures [54,55], has been described elsewhere [11]. The deposition frequency was varied from 1 to 6 Hz, so that the resultant thickness of the films was always approximately 20 nm.

### 2.2. Measurement and microscopy techniques

Magnetization measurements were carried out on Quantum Design vibrating sample magnetometer physical properties measurement system (VSM PPMS) with the magnetic field applied perpendicular to the surface of the films. For magnetization dependence on applied magnetic field measurements magnetic field sweep rate was kept at 5.0 mT/s. The frequency of VSM was 40 Hz and the amplitude of vibrations was 2 mm for YBCO samples and 1 mm for FePt samples. The dependence of YBCO measured superconducting properties on VSM settings and magnetic field sweep rate can be found in Refs. [56,57]. The $J_c$ of YBCO films was determined from the magnetization measurements using Bean formula for rectangular samples [58]: $J_c = 2\Delta M/[w_p(1 - w_p/3l_p)]$ in A/m$^2$, where $w_p$ and $l_p$ are respectively width and length of the samples measured, $\Delta M$ is the opening of hysteresis loops per unit volume. For YBCO films, $l_p = w_p = 5$ mm.

Atomic force microscopy (AFM) imaging of YBCO films was performed using an MFP-3D Asylum AFM (Asylum Research) in AC mode using Al reflex coated silicon probes with approximately 330 kHz resonance frequency and 42 N/m spring constant (NCHR