# A Flux Screening Technique for Growth of High-Quality Ferrite Films by Pulsed Laser Deposition

Yajie Chen[1], Tao Lu[2], Xiaoyu Zhang[2], Carmine Vittoria[1], *Fellow, IEEE*, and Vincent G. Harris[1]

[1]Center for Microwave Magnetic Materials and Integrated Circuits, Department of Electrical and Computer Engineering, Northeastern University, Boston, MA 02115 USA
[2]Department of Physics, Suzhou University, Suzhou, Jiangsu 215006, China

**We present a new technique to improve growth of ferrite films by pulsed laser deposition (PLD). The technique utilizes a metal screen placed between target and substrate, effectively to impede formation of large particulates on the surface of ferrite films by changing the plume shape and plasma density in the PLD chamber. Structural, magnetic, electrical, and microwave properties were measured for the flux screened and conventionally grown films. We demonstrate that the flux screen deposited MnZn-ferrite film has a lower coercivity (39 versus 58 Oe) and ferromagnetic resonance linewidth (350 versus 430 Oe at 9.58 GHz). The improvement in magnetic properties is associated with a reduction in large particulate contamination. This simple approach has the potential to produce high-quality ferrite films for use in electronic and microwave devices.**

***Index Terms*—Ferrite, ferromagnetic resonance (FMR), pulsed laser deposition (PLD).**

## I. Introduction

THE MnZn ferrite films are promising candidate materials for use in high-frequency electronic devices, due to high permeability and high resistivity. Since modern high-frequency device technology is shifting toward planar miniaturization, ferrite films may play an important role in facilitating the design and fabrication of devices such as micro-inductors, microtransformers, and microwave nonreciprocal devices [1]. The pulse laser deposition (PLD) technique is well suited for ferrite film fabrication due to the ability to ablate multicomponent materials stoichiometrically and to produce an energetic flux of atoms and ions. One drawback, however, is the fact that large particulates tend to be generated from the target material and become embedded in the growing film. These particulates, which range in size from $<0.1$ to several microns in diameter, can destroy the integrity of multilayer films, produce film layers with very rough surfaces, and make the patterning of small features difficult [2]. Over the past 20 years, many approaches have been proposed to minimize these large defects, including the use of reduced laser beam fluence [3], off-axis laser ablation [4], velocity filters [5], and the crossed-flux technique [6].

## II. Experiment

In this paper, we propose a scheme for the reduction of the density of large particulates in PLD grown ferrite films. In this approach, a stainless steel screen is placed between the MnZn-ferrite target and the substrate in a traditional PLD system as depicted in Fig. 1. The screen was a #400 mesh with a grid spacing of $\sim 30\ \mu$m. The area of the screen is large enough to intercept the entire plume in front of the substrate. MnZn ferrite films were deposited on (400) Si substrates by ablating a stoichiometric $Mn_{0.48}Zn_{0.46}Fe_{2.06}O_4$ target prepared by conventional ceramic processing. A KrF excimer laser operating at 5 Hz and 1.5–2 J/cm$^2$ was used in film processing. After evacuation to a base pressure of $2 \times 10^{-3}$ Pa, a dynamic pressure of argon gas ranging from 0.1 to 20 Pa was attained. The deposition process was carried out at a substrate temperature 600 °C over a period of 1–2 h.

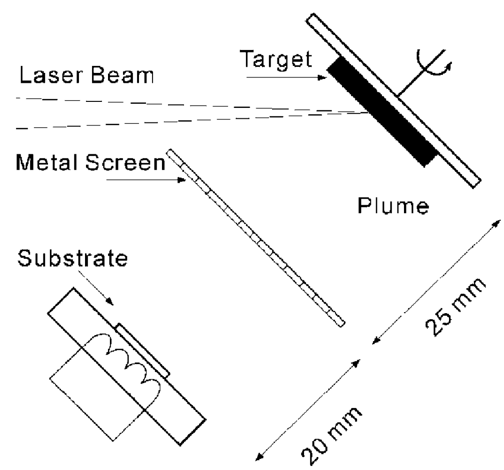


Fig. 1. Schematic diagram of PLD chamber with flux screen in place.

The phase, structure, and morphology of the films were determined by X-ray diffraction (XRD) and scanning electron microscopy (SEM). The composition was determined by energy dispersive X-ray spectroscopy (EDXS). We also measured magnetic hysteresis in the plane of the films at room temperature using a vibrating sample magnetometer (VSM). The film thickness, determined by surface profilometry, ranged from 90 to 500 nm for samples grown at different conditions. Electrical resistivity was measured using a standard four-probe method.

Digital Object Identifier 10.1109/TMAG.2006.879899

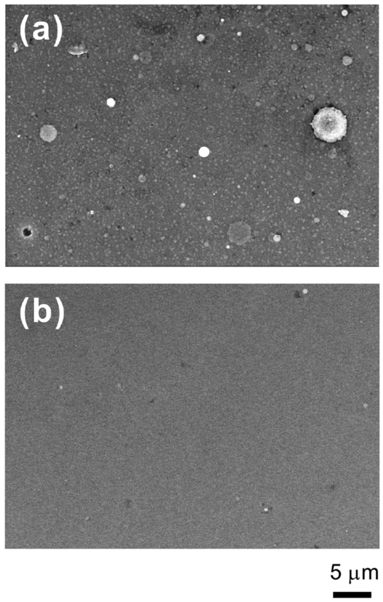


Fig. 2. Scanning electron micrographs illustrating surface morphology of MnZn ferrite films produced by (a) conventional and (b) flux-screened PLD techniques.

Although many samples have been prepared and characterized in order to explore the role of the flux screen in the structure and magnetism of the films, we chose to compare and contrast two films that are representative of the conventional and flux-screened growth techniques.

## III. Results and Discussion

All samples were determined to have a pure phase spinel MnZn-ferrite structure by XRD. Fig. 2(a) and (b) shows the surface morphology of a typical PLD film sample (i.e., no flux screen) and a film deposited with the flux screen in place, respectively. As is clearly illustrated in Fig. 2(a), the normal film contains many particulates which range in size from 0.2 to 5 $\mu$m. Usually, either liquid or solid ejecta tend to form particulates in size ranging from micron to submicron, as well as spherical and/or irregular shapes. It has been suggested that the particulates may arise from the interaction between the laser beam and the target as well as the plasma flux interaction with the background gas and substrate [2].

EDXS analysis indicates that the chemistry of the particulates was the same as the underlying film, suggesting that the particulates originate from the interaction between laser and target. However, the use of the screen results in a dramatic decrease in the particulate density as is seen in Fig. 2(b). Clearly, the film exhibits a smoother surface with very few large particulates. We suggest that the screen provides two mechanisms for reducing particulate density. One obvious mechanism is to physically obstruct the particulates larger than the 30-$\mu$m pore size of the screen. A second mechanism, which is less obvious but perhaps more important, is the role of the screen in adjusting the shape and particulate distribution of the plume. We believe that the electrically grounded screen alters the plume shape by changing the electrostatic field acting on the molecular flux and the particulates of the plasma. This assumption has been substantiated by

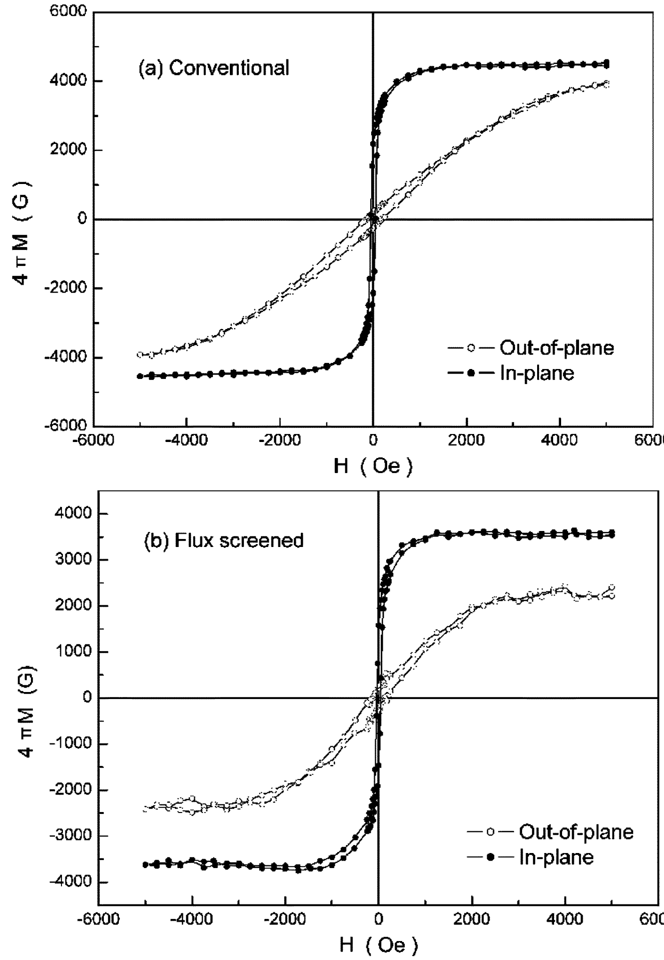


Fig. 3. Magnetic hysteresis loops of MnZn ferrite films produced by (a) conventional and (b) flux-screened PLD techniques.

a direct observation of the plume shape and plasma density. Actually, this action leads to an increase in collisions: 1) between the particulates and the screen grids, and 2) between particulates near the screen. A deleterious effect of the screen is a reduction in the throughput of molecular and atomic species leading to a 30%–50% reduction in the deposition rate. It, nonetheless, provides a marked improvement in the surface quality of the ferrite films and has been shown to allow film growth with high laser power and at low substrate temperatures [7].

Hysteresis loops of the normal and flux screened samples are very similar, except for a reduction in saturation magnetization for the screened film. Fig. 3 shows hysteresis loops for the samples deposited under an argon pressure of 2 Pa. The conventional sample has an average grain size of $\sim$300 nm and a coercive field of 58 Oe in the film plane; whereas, the flux screen film exhibits smaller grains of $\sim$80 nm and a coercivity of 39 Oe. These grains are small enough to form single domain particles since MnZn ferrite of the composition studied here has a critical single domain size of 3.8 $\mu$m [8]. The reduction in $H_c$ is predominately due to a decrease in domain wall pinning centers such as pinholes and particulates that are present in greater numbers in the film grown by conventional PLD.

The saturation magnetization exhibits a decrease from $4\pi M_s = 4500$ to 3500 G. Electrical measurements indicate that

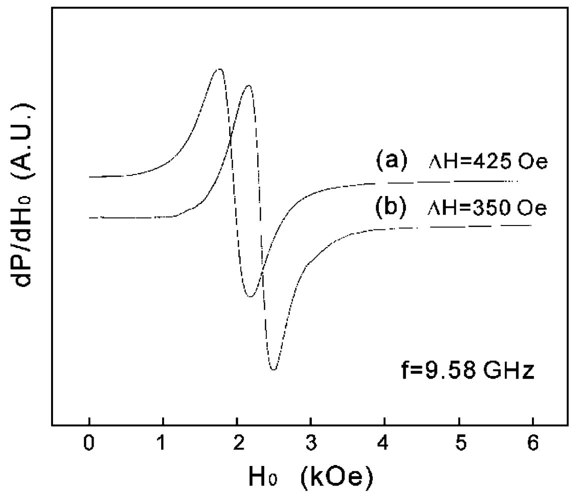


Fig. 4. FMR absorption of MnZn ferrite films produced by (a) conventional and (b) flux-screened PLD techniques.

the flux screened sample has an order of magnitude higher resistivity than that of the conventional sample. These results may be attributed to an increase in grain boundary density due to the fine-grained microstructure observed in the flux screened film and to the interface between the substrate and film [9]. Additionally, it may be assumed that the reduction in magnetization is associated with cation distribution on the A and B sublattice (i.e., Mn ion) in the spinel ferrite structure. We speculate that the change in plume shape and plasma density leads to an increase in ion collision frequency as they traverse the space between target and substrate. This leads to a reduced kinetic energy of Mn ions as they arrive at the substrate. This reduced kinetic energy, in turn, reduces surface mobility and leads to the freezing in of cation disorder and an increase in Mn inversion. This assumption is supported by the work of Yang *et al.* [10], which indicated PLD-induced Mn inversion in Mn-ferrite was directly correlated with a significant reduction in magnetization.

FMR measurements were carried out using a $TE_{102}$ mode at X-band frequencies. The conventional film has a derivative FMR linewidth, $\Delta H$, of 425 Oe at $H_0 = 1965$ Oe with external field aligned parallel to the plane of the film at 9.58 GHz. In contrast, the flux screened sample has a $\Delta H = 350$ Oe at $H_0 = 2216$ Oe. This linewidth is significantly lower than previous reports of linewidths in polycrystalline MnZn ferrite films ($\Delta H = 530$–$1600$ Oe) [11]. The decrease in linewidth results mainly from the smooth surface of the ferrite film. We estimated the $g$ factor to be 1.922 and 1.933 for conventional and flux screened samples, respectively. In doing so, we assumed the FMR equation: $\omega = \gamma\sqrt{H_0(H_0 + 4\pi M_s)}$ in an applied field parallel to the film plane. These deduced $g$ factors are comparable to some values reported in the literature for bulk MnZn ferrite [12]. Compared to other reported values, they are slightly smaller, in which case the reduction is attributed to an excessive iron composition that is also measured in our samples [13]. We believe that further narrowing of the linewidth can be achieved by raising the deposition temperature, thus reducing the grain boundary density.

## IV. Conclusion

In summary, a significant reduction in particulate density and size was demonstrated by positioning a metal screen between the target and the substrate in a traditional PLD chamber. Structural, magnetic, electrical, and microwave properties were measured for flux-screened and conventionally grown films. We demonstrate that the flux screened deposited MnZn-ferrite film has a lower coercivity and ferromagnetic resonance linewidth (350 Oe at 9.58 GHz) due to a higher quality morphology. This simple approach has the potential to produce high-quality ferrite films for use in electronic and microwave devices.

## References

[1] H. L. Glass, "Ferrite films for microwave and millimeter-wave devices," *Proc. IEEE*, vol. 76, pp. 151–158, 1988.

[2] D. B. Chrisey and G. K. Hubler, *Pulsed Laser Deposition of Thin Films*. New York: Wiley, 1994.

[3] A. Gupta, B. W. Hussey, A. Kussmaul, and A. Segmüller, "Defect formation caused by a transient decrease in the ambient oxygen concentration during growth of $YBa_2Cu_3O_7$ films," *Appl. Phys. Lett.*, vol. 57, pp. 2365–2367, 1990.

[4] R. J. Kennedy, "A new laser ablation geometry for the production of smooth thin single-layer $YBa_2Cu_3O_{7-x}$ and multilayer $YBa_2Cu_3O_{7-x}$/$PrBa_2Cu_3O_{7-x}$ films," *Thin Solid Films*, vol. 214, pp. 223–228, 1992.

[5] D. Lubben, S. A. Barnett, K. Suzuki, S. Gorbatkin, and J. E. Greene, "Laser-induced plasmas for primary deposition of epitaxial Ge and Si films," *J. Vac. Sci. Technol. B*, vol. 3, pp. 968–974, 1985.

[6] M. D. Strikovsky, E. B. Klyuenkov, S. V. Gaponov, J. Schubert, and C. A. Copetti, "Crossed fluxes technique for pulsed laser deposition of smooth $YBa_2Cu_3O_{7x}$ films and multilayers," *Appl. Phys. Lett.*, vol. 63, pp. 1146–1148, 1993.

[7] M. Nakano, M. Akasea, H. Fukunaga, Y. Matsuob, S. Yabukami, M. Yamaguchic, and K. I. Arai, "Permeability in PLD-made Mn–Zn ferrite thin films by low-temperature process," *J. Magn. Magn. Mater.*, vol. 242-245, pp. 157–159, 2002.

[8] J. Aarts, I. A. Shiekah, and P. J. van der Zaag, "Domain structure in polycrystalline MnZn ferrite imaged by magnetic force microscopy," *J. Appl. Phys.*, vol. 85, pp. 7302–7309, 1999.

[9] Y. Hoshi, Y. Kubota, and H. Ikawa, "Preparation of c-axis-orientated barium ferrite thin films with small crystallite size," *J. Appl. Phys.*, vol. 81, pp. 4677–4679, 1997.

[10] A. Yang, X. Zuo, C. Vittoria, and V. G. Harris, "Magnetism, structure and cation distribution in $MnFeO_4$ films processed by conventional and alternating target laser ablation deposition," *IEEE Trans. Magn.*, vol. 42, no. 10, pp. 2870–2872, Oct. 2006.

[11] Q. Y. Yan, R. J. Gambino, S. Sampath, and Q. Huang, "Neutron diffraction and ferromagnetic resonance studies on plasma-sprayed MnZn ferrite films," *J. Appl. Phys.*, vol. 97, p. 033 902, 2005.

[12] S. C. Byeon, K. S. Hong, and I. T. Kim, "Line width of manganese-zinc ferrite polycrystals with oxygen partial pressure," *J. Appl. Phys.*, vol. 83, pp. 6873–6875, 2005.

[13] W. A. Yager, J. K. Galt, and F. R. Merritt, "Ferromagnetic resonance in two nickel-iron ferrites," *Phys. Rev.*, vol. 99, pp. 1203–1210, 1955.

Manuscript received March 2, 2006 (e-mail: y.chen@neu.edu).