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Fully magnetic printing by generation of magnetic droplets on demand with a coilgun

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In this paper, we exhibit a coilgun-based approach to drop-on-demand printing of liquids laden with magnetic particles. In contrast to other drop-on-demand technologies designed to print droplets only in gaseous environments, this methodology allows one to print magnetic droplets inside any gaseous or liquid media using the same coilgun. Furthermore, we demonstrate the basic principles of magnetic drop-on-demand generation and show the physico-chemical parameters controlling the process. © 2015 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4937152>]

I. INTRODUCTION

Drop-on-demand (DOD) technology is used in different engineering and analytical applications.^{1–3} Materials science and engineering applications, including additive manufacturing (3D printing), require formation and printing of complex materials, magnetic in particular.^{4–10} Since the beginning of the inkjet printing era, a long standing challenge of fully magnetic printing has been to break up magnetic fluids into droplets when the fluids are being pulled from a nozzle by a magnetic field.¹¹ It appears difficult to form droplets of magnetic fluids on demand by applying a magnetic field primarily because magnetic fluids always form a long liquid bridge connecting the drop with the nozzle.¹² The jetting of magnetic fluids is, therefore, considered as an obstacle for the realization of the fully magnetic DOD. On the other hand, in contrast to other DOD technologies designed to print droplets only in gaseous environments, magnetic DOD technology would allow one to print magnetic droplets inside any gaseous or liquid media; henceforth, the technology is very attractive.

Recently, interest on the printing of magnetic materials was rekindled by the opportunity to control the alignment of magnetic particles on the target.^{4,6,10} It appears that by using available printheads, one can produce droplets of magnetic fluids on demand and then manipulate them with a magnet positioned close to the target. This way, magnetic particles could be aligned anisotropically in square and circular shaped magnetic films. Magnetically patterned samples show significantly increased high frequency permeability along with decreased hysteresis losses as compared to the test samples that were not magnetically aligned.¹⁰ Thus, printing of magnetic materials opens up opportunities in manufacturing magnetic composite materials, inductors, and antennae.

In this paper, we present an experimentally validated approach, which resolves the long-standing challenge of fully magnetic printing of fluids loaded with magnetic particles. It is demonstrated that the droplets can be pulled out from the reservoir by the applied magnetic field without any need for a hydrodynamic push or any other mechanical agitation of the liquid surface.

II. EXPERIMENTAL METHODS

The basic idea of the proposed method is as follows. In droplets loaded with magnetic particles, the magnetic moment is proportional to the droplet volume V and magnetization $\mathbf{M}$ as $\mathbf{m} = V\mathbf{M}$. Assuming a linear constitutive equation for the magnetization versus field, $\mathbf{M} = \chi\mathbf{H}$ where χ is the magnetic susceptibility, we have $\mathbf{m} = \chi V\mathbf{H}$. Thus, the driving force acting on the drop is written as $\mathbf{F}_m = \mu_0\chi V(\mathbf{H} \cdot \nabla)\mathbf{B} = \chi V(\mathbf{B} \cdot \nabla)\mathbf{B}/\mu_0$, where μ_0 is the permeability of vacuum and $\mathbf{B}$ is the induction vector. This force significantly depends on the field gradient; therefore, it is critical for magnetic printing to generate a strong field gradient.⁸ In order to satisfy this requirement, we employ a coilgun as the field generator.

A coilgun with an iron core is the main driving element of the proposed magnetic DOD, see Figure 1. To break up the liquid bridge following the droplet, one of the greatest challenges in the design of any drop-on-demand generator,^{1,13} we used tungsten wire as a droplet holder. Having a wire as a support for the droplet is favorable for the DOD operation because this wire naturally creates a counter capillary pressure supporting wire dewetting and prevents the feeding of a long tail behind the moving drop.¹⁴ Tungsten wire of 125 μm in diameter was attached to the XYZ linear translation stage (Thorlabs PT3). The wire axis was coaxially

FIG. 1. (a) Schematic of the magnetic DOD platform: 1–XYZ Stage, 2–Holder, 3–Drop, 4–Substrate, 5–Iron Core, 6–Coil, 7–Switch, 8–Power Supply. Drop-on-demand generation of (b) ferrofluid droplets under water (with satellite) and (c) MIEX resin under water.

aligned with the coil axis. A drop was delivered to the wire surface either manually or using a syringe pump.

The coil was made of AWG36 copper wire and had the total resistance of 1 k Ω ; it was connected to the power supply (TDK Lambda ZUP 120-1.8) controllable through the PC. A piece of paper was placed atop of the coilgun and was attached to the XYZ translation stage. To visualize the process of drop on demand generation, a fast framing camera (IDT MotionPro) coupled with a macroscopic objective was used. To assure the best contrast of the recorded frames, a backlight illumination was set up by using a light source (Chiu Technical Corp., F0-150) with flexible arms.

Typical voltage and current waveforms used to generate DOD are shown in Figure 2(a).

In order to eliminate the effect of temperature increasing the copper wire resistivity, the power supply was operated in the current limiting mode. This mode allowed one to keep the current constant [see Figure 2(a)] with fluctuations not exceeding 4/10 mA. The rise time of the current pulse was about a half second as illustrated in Figure 2(b).

An axial component of the magnetic field generated by the coilgun was measured by the linear Hall-effect sensor (BCD Semiconductor Manufacturing Limited, AH49E). An output sensitivity of the Hall-effect sensor is found to be 1.77 mV/Gauss. The absolute calibration of magnetic field was carried out using the Hall-effect digital teslameter DTM-133 with the attached head MPT 132 7S. This set of measurements allowed us to study the axial and radial magnetic field shown in Figures 2(c) and 2(d), respectively. The axial field can be approximated (using OriginLab's OriginPro) by the formula

$$B(x) = \frac{B_0}{(1 + ax^{b-1})}, \quad (1)$$

where B_0 is a maximum field magnitude measured at the coil edge and defined by the coil current, x is a distance from the

coil edge, and constants a and b depend on the coil geometry, but do not depend on the current that we pass through the coil. Along the symmetry axis, the radial component of magnetic field is zero. As discussed in Ref. 8, in the vicinity of the central axis, the radial component of magnetic field creates a self-focusing effect—small radial deviations of the flying drop are suppressed by the radial component of magnetic force pushing the drop toward the axis.

By passing lower currents through the coil and measuring the magnetic field, we confirmed that the field distribution follows the same formula (1), provided that the only constant B_0 is adjusted accordingly. For example, for a current of $I = 86$ mA, the constant B_0 is equal to $B_0 = 950$ G. This analysis suggests that the current completely controls the coilgun performance and Eq. (1) provides a convenient analytical approximation of the field distribution. The obtained field distribution is also favorable for DOD operations since the small radial component of the magnetic field helps to suppress minute deviations of the drop trajectory from the straight line.

One notices that the strength of a magnetic field reaches its maximum at the coil edge. However, the field gradient in the axial direction reaches its maximum about 2 mm away from the coil edge; see Figure 2(c). This distinct behavior between the magnetic field and its gradient has to be taken into account because the field gradient controls the drop acceleration. As follows from the non-uniform distribution of the field gradient, the drop acceleration is not constant along its pathway to the target. To approximate the axial component of magnetic force, we substituted Eq. (1) into the formula $F_m = \chi V(B_x \cdot (dB_x/dx))/\mu_0$ and fit the resulting equation with OriginPro[®] to get

$$F_m = F_0 \exp\left(-\frac{x}{L_H}\right) \sin\left(\pi \frac{x - x_0}{w}\right), \quad (2)$$

where parameters $L_H = 9.47 \times 10^{-4}$ m, $x_0 = 22.1$ m, and $w = 11.1$ m depend only on parameters a and b in original

FIG. 2. (a) The voltage and current waveforms used in the DOD coilgun. (b) The rise time of the current in the coilgun. (c) Measured (black) and approximated (red) axial component of the magnetic field and its gradient (blue) generated by the coilgun with the current of 86 mA. (d) Radial component of magnetic field measured at 20 mm distance away from the coil edge.

Eq. (1) and prefactor F_0 depends on B_0 and drop parameters χ and V . The actual magnitude of the axial component of magnetic force is defined by the coil current and is accounted for in Eq. (2) by the F_0 parameter. Since it depends on the drop size, below we consider this parameter as an adjustable constant.

III. RESULTS AND DISCUSSION

A. Stages of drop detachment

Magnetic droplets of different volumes were used to demonstrate the robustness of magnetic DOD. Figure 3 illustrates the process of magnetic printing where a ferrofluid drop (FerroTec EFH1, volumetric density of magnetic particles is 7.9%) was withdrawn from a tungsten wire (on the right) by the coilgun (on the left).

The operation of the magneto-generator can be divided into 4 stages corresponding to different physical phenomena. Upon passing the current through the coilgun, the drop sitting on the wire (i) first slides over the wire to its tip, (ii) then it elongates and gets broken and detached from the wire, (iii) then gets caught by the field gradient and accelerates and moves toward the target, and (iv) impinges on the target. Using a drop-tracking algorithm implemented in Matlab[®] (its basic principles are described elsewhere^{14,15}), we reconstructed the drop trajectory. It was observed that the drop slides towards the wire tip with almost constant velocity and leaves the wire at some finite velocity. For a particular experiment shown in Figure 3, each (i)–(iii) process takes on its own time interval which is given in (b). A small bend of the drop trajectory around the 15th ms in (b) reflects the formation of a pendant drop.

When a drop has been formed, it flies toward the target with acceleration. The drop velocity reaches its maximum near the edge of the coilgun. For example, the maximum velocity of the drop shown in Figure 3 is estimated as $v = 2.5$ m/s. Upon impact, the drop deforms and its visible cap dimples and bulges a few times before the drop finds its equilibrium configuration without visible disintegration.¹⁶ In

the case of porous targets, such oscillations cause flow pulsations and, hence, the rate of penetration of droplets into pores depends on these oscillations. This leads to a nonconventional mechanism of the drop penetration involving the coupling with magneto-hydrodynamics.

B. Versatility of coilgun DOD technique

When using the developed coilgun DOD generator, one can print different patterns by moving the substrate.^{14,17} As an illustration, “CU” has been printed on a Whatman qualitative filter paper (grade 4) by moving this paper over the edge of the coilgun, Figure 4. The droplets spread over the paper and partially invade the pores; hence the borders of the droplets are not smooth.

The droplets were guided to hit the intersections of the grid with 1 mm spacing. An average diameter of the printed pixel was 1 mm (Figure 4(b)).

C. Drop flight analysis and modeling

The coilgun appears to control the droplet placement fairly well. In order to demonstrate its performance and accuracy, we traced the drop trajectory and varied the droplet size. In the Cartesian system of coordinates, with the center at the wire end and the x -axis along the wire centerline, the drop flight was modeled by the Newton's equation of motion

$$m \frac{d^2x}{dt^2} + \alpha S \left(\frac{dx}{dt} \right)^2 = F_m, \quad (3)$$

where $x(t)$ is the drop position along the x -axis, m is the drop mass, S is its cross-sectional area, and F_m is the magnetic force defined by Eq. (2). Parameter α is introduced as follows $\alpha = C_{shape} \rho_{air}$, where C_{shape} is a drag coefficient¹⁸ and ρ_{air} is the air density. The initial position of the drop was taken at the center of coordinates located at the wire edge and the initial velocity of the drop v_0 is considered known.

However, the drop velocity just after its detachment is difficult to measure with high accuracy due to a very fast and

FIG. 3. (a) A set of frames illustrating the drop detachment and impact on the target. Exposure time is $4.9 \mu\text{s}$, frame rate is 2000 fps, and air gap thickness is 4 mm. (b) Trajectory of the droplet center of mass and (c) velocity profile.

FIG. 4. (a) Letters “CU” printed by magnetic DOD generator on the Whatman paper (Grade 4). (b) Microscope image of a single drop sitting on the Whatman paper. (c) Typical experimental (solid lines) plots of drop trajectories and corresponding theoretical (markers) trajectories. Legend of the drops is shown in Table I.

complicated set of dynamics in the detachment process. It was also impossible to make any temporal correlations between the drop position and the strength of the magnetic field at this position. As shown in Figure 2(b), the rise time of the current in the coil is about 0.5 s, whereas the drop detachment and flight take only milliseconds. Therefore, the drop may have already taken off and been flying while the coil current is still on the rise and, thus, it never reaches the plateau of its maximum amplitude. In order to combat these uncertainties, we used initial velocity v_0 and parameter F_0 as adjustable factors.

It is convenient to introduce dimensionless variables, $X = \frac{x}{L_H}$, $T = \frac{t}{t_0}$, where the characteristic length L_H is defined in Eq. (2). The characteristic time t_0 was introduced as $t_0 = L_H/v_0$. All parameters used in Eq. (3) are defined in Table II. Rewriting Eq. (3) in the dimensionless form, we have

$$X'' + \beta X'^2 = \gamma \exp(-X) \sin\left(\pi \frac{L_H}{w} [X - x_0/L_H]\right),$$

$$X(0) = 0, X'(0) = 1, \quad (4)$$

where $\beta = \frac{\alpha SL_H}{\rho_f V}$ and $\gamma = \frac{F_0 L_H}{\rho_f V v_0}$. It is clear that solution of Eq. (4) depends only on the ratio of F_0/v_0 of unknown parameters. Numerically generating solutions of Eq. (4) $x_i^{th}(v_0, F_0)$ at i -th time moment with different parameters γ and β , one can fit the observed trajectory by numerically minimizing the quadratic functional $\Psi = \sum_i (x_i^{\text{exp}} - x_i^{th})^2$ for a set of experimental data points x_i^{exp} taken at the image frame i . The results of this fitting procedure are shown in Figure 4(c). In spite of large variations in the drop volume and gap distance, the kinetics of all drops detached from the substrate can be described by Eq. (3).

As follows from the definition, magnetic force depends on the environmental medium via magnetic susceptibility χ only ($\chi \cong 1$ for air and does not appear in equation), whereas a counteractive force may include additional terms depending on the medium. To show the universality of the magnetic DOD generator and its applicability in various applications, the drops were also generated in a denser medium—water. Underwater drop-on-demand generation was carried out with the same coilgun and ferrofluid (shown in Figure 1(b)). Besides ferrofluid, another fluid with magnetic particles (MIEX[®] resin) and various magnetic nano- and micro-particles were also tested with the magnetic generator. In Figure 1(c) we demonstrate the manipulation of MIEX clusters used for magnetic ion exchange technology of cleaning water.¹⁹ These beads are initially held together by a very

TABLE I. Parameters of the drops listed in Figure 4(c).

Drop number	Gap distance, (mm)	Drop volume, 10^{-12} (m ³)	Initial velocity, (m/s)	Prefactor in Eq. (2) F_0 , (μ N)
1	2.4	28	0.55	227.2
2	4.8	159	1	2321
3	4.5	60	0.9	950.6
4	5.2	192	0.96	2811
5	3.4	34	0.66	318.2

TABLE II. Parameters used for calculations.

Parameters used for calculations			
Variable	Value	Unit	Description
Magnetic field fitting			
a	0.17504	1/m	Fitting parameters
b	2.72397	Dimensionless	
B_0	Adjustable	Tesla	Maximum field strength
Parameters of ferrofluid and environment			
σ	0.03	N/m	Surface tension of ferrofluid
μ_0	1.25664×10^{-6}	N/A ²	Permeability of a vacuum
χ	2.64	Dimensionless	Ferrofluid magnetic susceptibility
ρ_f	1.21×10^3	kg/m ³	Ferrofluid density
Setup parameters			
d	125×10^{-6}	m	Diameter of wire-holder
Magnetic Force Parameters			
x_0	22.11524	m	Defines the profile of the magnetic force near the coil edge
w	11.05763	m	
L_H	9.47×10^{-4}	m	Characteristic length of magnetic force decay
F_0	Adjustable	N	Magnitude of magnetic force
Others			
v_0	Adjustable	m/s	Droplet velocity just after detachment

small interfacial tension and dipole-dipole forces. When the field is applied, a few MIEX beads can be separated from the cluster and pulled away by the magnetic force. This example illustrates the possibility of printing very complex materials that cannot be considered fluids.

D. Effect of the coilgun parameters on the drop size

In Secs. III A–III C, we consider a particular embodiment of the coilgun idea and used the measured parameters of the coil/droplet pairs to illustrate the robustness of the developed DOD coilgun generator. It is instructive to discuss the effect of coilgun parameters on the droplet size, assuming that the current can be increased and the droplets can be made of different materials. Therefore, we want to establish a criterion for the drop detachment considering the holder diameter d , the strength of magnetic field B_0 , and susceptibility χ as the main variables of interest. The criterion is based on the assumption that the drop of volume V will be detached from the wire of diameter d when the magnetic force is greater than the wetting force F_w , see Figure 5. This force is acting over the wire perimeter, and when the drop is about to leave the wire, the force is estimated as $F_w = \pi \sigma d$, where σ is the surface tension of the drop.²⁰

Thus, the drop detachment criterion is set as follows

FIG. 5. Force balance criterion when drop is about to leave from the wire holder.

$$\pi d \sigma \leq \frac{\chi V B_x (dB_x/dx)}{\mu_0} \text{ or } V \geq V_{\min} = \frac{\pi d \sigma \mu_0}{\chi B_x (dB_x/dx)}. \quad (5)$$

This criterion implies that the droplets larger than the right-hand side of inequality (5) can be detached from the holder. Employing approximation (2), this criterion can be calculated using parameters from Table II. Analysis of Eq. (5) reveals several approaches to reduce the extracted drop volume and, thus, to improve the printing resolution. Since magnetic field generated by the coil is proportional to the current I , the denominator of Eq. (5) strongly depends on the current, $B_x (dB_x/dx) \propto I^2$. Therefore, the coil current can effectively control the printed drop volume and it defines the minimum drop volume being extracted by a particular setup.

The radius of the minimal drop that can be printed by the given coilgun significantly depends on the drop position with respect to the edge of the coilgun. This dependence of the drop radius on the coilgun position is shown in Fig. 6 for different $B_0 \chi^{1/2}$.

As follows from Fig. 2(c), the field gradient reaches a maximum value when one moves away from the coilgun edge. Accordingly, the magnetic force reaches its maximum value not at the coilgun edge, but at a certain distance away from it. Therefore, one option to improve the printing resolution with the existing coilgun design is to increase magnetic field, i.e., to increase the current and take advantage of this distribution of the force field allowing one to print small droplets by choosing the right spacing between the coilgun and the droplet holder. For example, by generating the field $B_0 = 0.3$ T, one would be able to decrease the radius of droplets of the same ferrofluid with $\chi = 2.64$ down to $34 \mu\text{m}$ by positioning the coilgun 0.9 mm away from the holder edge.

Another option to improve the printing resolution with the existing coilgun design is to decrease the holder diameter d , i.e., the numerator of Eq. (5). This effect was verified by printing droplets from a wire of diameter $d = 78 \mu\text{m}$, which is about half of the diameter used in other trials shown in Figures 1 and 3. With this decrease of the wire diameter, we were able to print the 2.4 nl droplets, shown in Figure 7.

The presented analysis suggests that the droplet size can be decreased by either decreasing the holder diameter or increasing the current in the coilgun. By carrying out both of these

FIG. 6. Field dependence of the drop radius on the coilgun position measured with respect to the holder edge.

FIG. 7. Magnetic printing of Ferrofluid EFH-1 from a $78 \mu\text{m}$ wire.

drop volume reduction methods, one could vastly improve the printing resolution of the coilgun-based DOD generator.

IV. CONCLUSION

This work shows how to defeat the jetting of magnetic fluids, which is the main obstacle for the realization of the fully magnetic drop-on-demand printing technology. There are two key ideas behind this technology. The first is the implementation of a coilgun which generates a special non-uniform magnetic field. As a result, this field generates a force profile with a maximum situated some distance away from the coil edge. Thus, on its way to the target, the drop is accelerated and breaks the liquid bridge connecting it to the holder. The second idea is to implement a cylindrical holder-wire in the drop delivering system. By using such a holder, the liquid film coating the wire cannot effectively supply the liquid to the liquid bridge. Hence, the bridge connecting the drop to the wire cannot be continuously fed, which leads to its breakup. The universal character of this method was proven by DOD generation in air and underwater by utilizing the same coilgun. Coilgun-based design of the DOD generator allows reliable, controllable, and predictable generation of drops of various dimensions and composition. This paper was merely a proof of concept that magnetic printing of drops-on-demand in multiple mediums is not only possible but also easily achievable. With further experimentation, the proposed magnetic printing method will expand the variety of DOD applications immensely, specifically in the fields of biology, medicine, microfluidics, and material science.

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