Ejection of ferrofluid grains using nonlinear acoustic impulses—A particle dynamical study

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We consider a model dilute ferrofluid with the grains suspended in water (e.g., γ-Fe_2O_3) and subject the system to a strong, homogeneous magnetic field directed perpendicular to the surface such that there is chain formation along the field direction. We show that an appropriate impulse initiated at the base of the container might travel as a nondispersive soliton pulse with sufficient energy to overcome surface tension and eject the ferrofluid grain nearest to the liquid–air interface. The proposed mechanism, if successfully realized in the laboratory, could help design a nozzle-free, ink-jet printer of unparalleled resolution. © *1999 American Institute of Physics.*
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Ferrofluids are colloidal systems that are composed of single-domain magnetic particles, which are usually coated with a dispersant molecular layer and are suspended in a nonmagnetic solvent.[1] Ferrofluid grains are, typically, ~10 nm in diameter. The present work is applicable to dilute ferrofluids in which ferrofluid grains can be approximated as elastic objects.[2]

Certain ferrofluid grains, such as γ-Fe_2O_3 ferrofluid grains which are ~8.5 nm in diameter, contain magnetic moments of about $2\times10^4\,\mu_B$.[3] When the ferrofluid grains can be suspended in water or oil and the system is subjected to a strong, homogeneous magnetic field B that is directed perpendicular to the ferrofluid surface, the ferrofluid grains tend to align in vertical chains. It turns out that γ-Fe_2O_3 ferrofluid grains can be suspended in water and one can make a stable ferrofluid.[3] Since water can be colored using dyes, γ-Fe_2O_3 ferrofluid and similar systems can potentially be used as inks. If one can design an ink-jet printer in which the sizes of the ink droplets are dictated by the sizes of the ferrofluid grains rather than by the nozzle diameters that inevitably limit the resolution of ink-jet printers, it may become possible to design ink-jet printers of unparalleled resolution. Such printers have the potential to be significantly faster than existing laser-jet printers and may be used for applications such as making small imprints to legitimize financial/commercial transactions and high-denomination currency bills to prevent counterfeiting.

The key obstacle to making a high-resolution ferrofluid printer lies in our inability to extract ferrofluid grains from the surface of a ferrofluid. No matter how strong the B field is and what its time dependence may be, it is not possible to extract ferrofluid grains from a ferrofluid.[4] In every case, it turns out that the droplets that one can extract are macroscopic in size. The field can be used to position the vertical chains of ferrofluid grains in the liquid (see Fig. 1). Thus, it is inconvenient and perhaps undesirable to use a time-varying field for extracting ferrofluid grains from a ferrofluid.

In this letter, we show via particle dynamical simulations that it is possible to preferentially evaporate ferrofluid grains aligned in chains from a dilute ferrofluid subjected to a uniform B field using *nonlinear acoustic pulses*.[5–12] Magnetic particles interact with each other via dipolar interactions $V(r_{ij})=-\Sigma_i\boldsymbol{\mu}_i\cdot\mathbf{B}+\Sigma_{i>j}(U_{ij}^{dd}+U_{ij}^{\mathrm{nm}})$, where $U_{ij}^{dd}=-(\mu_0^2/r_{ij}^3)[\boldsymbol{\mu}_i\cdot\boldsymbol{\mu}_j-3(\boldsymbol{\mu}_i\cdot\mathbf{r}_{ij})(\boldsymbol{\mu}_j\cdot\mathbf{r}_{ij})]$, and $U_{ij}^{\mathrm{nm}}=\epsilon[\exp(-[r_{ij}-d]/\eta-\exp(-[r_{ij}-d]/2\eta)]$.[13,14] We define, $r_{ij}\equiv|\mathbf{r}_i-\mathbf{r}_j|$ above, the first term on the right side of the expression for $V(r_{ij})$ describes the fact that the magnetic moments $\boldsymbol{\mu}_i=\mu_0\boldsymbol{\mu}_i$ (where $\boldsymbol{\mu}_i$ and $\mathbf{r}_{ij}$ denote appropriate unit vectors) of the ferrofluid grains tend to align along the field direction to minimize the potential energy of the system. The second term, U_{ij}^{dd} describes the interaction between

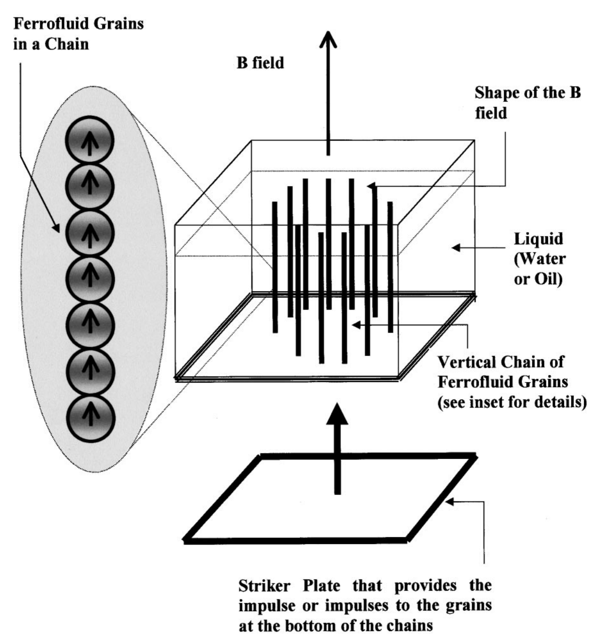

FIG. 1. Schematic showing chains of ferrofluid grains in a ferrofluid. The chains are aligned as dictated by the shape of the B field. The striker plate shown at the bottom generates the impulse.

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two ferrofluid grains or dipoles in the ferrofluid system. The term U_{ij}^{nm} describes the nonmagnetic interaction between the ferrofluid grains due to a soft-core repulsion at very short distances and a weak short-range attraction. We define $d=2R$, where R is the ferrofluid grains radius. In addition, if and when the ferrofluid grains touch one another, as can be the case when a strong, homogeneous B field is applied to the system, it is likely that there would be a repulsive force that would come into play. If the ferrofluid grains can be regarded as elastic objects, this force would be Hertzian[15] with the potential as described below:

$$V(\delta_{i,i+1})=(2/5D)(R/2)^{0.5}\delta_{i,i+1}^{5/2}\equiv a\,\delta_{i,i+1}^{5/2},\qquad \delta_{i,i+1}=d-(r_i-r_{i+1}), \tag{1}$$

where $D=(3/2)[(1-\sigma^2)/Y]$ and Y and σ refer to the Young's modulus and the Poisson's ratio of the material that make up the ferrofluid grains.

The typical numbers used are as follows: $d=10^{-8}$ m, ferrofluid grain mass $=2.72\times10^{-21}$ kg, $\mu_0=2.1\times10^4\,\mu_B$ the Bohr magneton. The energy of the dipolar interactions between the neighboring ferrofluid grains separated by d is given by $\mu_0^2/d^3\approx7.50\times10^{-18}$ J. We choose $\eta=2.5\times10^{-10}$ m and $\epsilon=1.28\times10^{-21}$ J.[13,14] In Eq. (1), we take $Y=1.0\times10^{11}\,\mathrm{N\,m^{-3/2}}$ and $\sigma=0$ (the precise values of Y and σ are unknown for most ferrofluid grains). An additional quantity of interest is the surface tension of water, which is $\approx7.3\times10^{-2}$ N/m. Gravity plays a negligible role, being about 2.67×10^{-20} N on each ferrofluid grain compared to the dipolar force $\approx2.28\times10^{-12}$ N. For ejecting ferrofluid grains through the water–air interface one must overcome a surface force of about 2.29×10^{-9} N. The Hertzian forces are $\approx10^{-8}$ N, and hence, dominate the dipolar forces.

At strong, vertical, homogeneous B fields (~200 G or 2×10^{-2} T), ferrofluid grains align in chains. The chains are vertical stackings of the ferrofluid grains with extremities at the base of the ferrofluid and at the water–air interface (see Fig. 1).[16] The nonmagnetic force can be ignored in dilute ferrofluids. The ferrofluid grains, which are dipoles, can be thought of as touching one another. We ignore thermal effects at strong magnetic fields.

We now address the problem of ejection of a ferrofluid grain by sending a nonlinear acoustic pulse from the bottom of the container to the surface of the liquid in the container, i.e., vertically upward along the direction of the applied B field. We contend that for appropriate magnitude of the initial impulse it is possible, to eject one ferrofluid grain from the surface into the air. We model a dilute ferrofluid in which the system has a large number of vertical ferrofluid grain chains (of about 500 ferrofluid grains) with extremities at the base of the ferrofluid container and at the surface of the ferrofluid. We focus on one of these chains (Fig. 1).

We note that the dipolar attraction between two adjacent ferrofluid grains in a vertical chain is $\propto 1/r^4$. The net effect of these forces is to allow the ferrofluid grains to touch one another. Gravitational force being negligible, the ferrofluid chains are not loaded. Thus, the centers of each particle are apart by d.

If an impulse is initiated at the bottom of the container (Fig. 1), we can state that $r_i(t)$ describes the displacement of each grain. We consider the Hertzian forces that come into play during pulse propagation and probe whether an impulse traveling along a chain can help overcome surface tension and eject at least one ferrofluid grain outside the liquid.

In equilibrium, there is no surface force on the top grain. When the top grain tends to move above the liquid surface, we assume that the force increases linearly with displacement from the equilibrium position, with the maximum value of the force being given by $2\pi R\gamma$, γ being the surface tension, which is the maximum static surface force that can be experienced by the macroscopic ferrofluid grain. We assume also that the "extraction work" which is required in order to remove one ferrofluid grain from the fluid is $\pi R^2\gamma$. Since the work done by the surface force should equal the extraction work, we can calculate an "escape distance." If the displacement of the ferrofluid grain exceeds this distance, the surface force becomes zero and the ferrofluid grain is "free." If the first ferrofluid grain escapes, the surface force is applied to the second ferrofluid grain and then one must determine whether the second ferrofluid grain possesses enough energy to escape, and so on.

The equation of motion of a spherical ferrofluid grain, in a chain of spherical grains in contact, labeled i at location r_i and moving with acceleration d^2r_i/dt^2, can be written as

$$md^2r_i/dt^2=k[(d-\Delta_0-r_i+r_{i-1})^{3/2}-(d-\Delta_0-r_{i+1}+r_i)^{3/2}], \tag{2}$$

where $k=5a/2$. The quantity Δ_0 gives the distance of closest approach between the grains in the absence of the pulse and is a parameter that describes the "loading" of the chain and is vanishingly small in this problem. The grain compression due to an impulse in this regime far exceeds the initial loading of the chain. As a result, an impulse imparted to an edge grain travels along the chain as a perfect soliton.[5–12] In studying our problem, we are interested in the latter regime.

We solve Eq. (2) numerically using the Gear algorithm.[17] The initial conditions are as follows: $dr_1/dt|_{t=0}=v_{\mathrm{init}}m/s$, $dr_i/dt|_{t=0}=0$ for $i>1$ where $i=1$ defines the grain that is nearest to the striker plate in Fig. 1. The soliton propagates from the bottom of the chain to the surface grain at the water–air interface at fixed velocity c. For ferrofluid grains of diameter 10^{-8} m, we find that $v_{\mathrm{init}}\geqslant67$ m/s for the surface grain to eject. Our calculations suggest that such initial velocities lead to forces that result in grain compressions of about 5%. The normalized velocity of ferofluid grains as a soliton passes through them is shown in Fig. 2 where the center of the soliton is placed at grain number 0, i.e., the kinetic energy of the soliton is concentrated within the central grain. The adjacent grains, while a part of the traveling soliton, carry a very small fraction of the total kinetic energy carried by the soliton.

Solitons are strongly nonlinear objects, and hence, their velocities are related to their amplitudes. Our simulations show that the soliton velocity $c\propto A^{1/4}$, where A is the amplitude of the displacement of a grain as a (primary or secondary) soliton passes through it.[10–12] Since no realistic system is ideal, it is important to account for dissipative effects. An important dissipative effect arises due to restitution associated with differences in the intergrain force as the grains compress (load) or decompress (unload). We define $w=F_{\mathrm{unloading}}/F_{\mathrm{loading}}<1$ if restitution is present. We find that

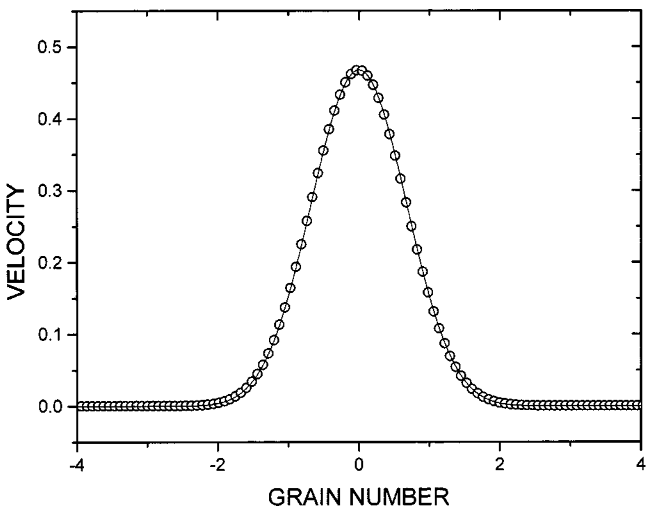

FIG. 2. Plot of normalized velocity of grains vs grain location as a soliton passes through the grains. The grains are numbered with the origin fixed at the center of the soliton. The maximum grain velocity depends upon v_{init}. We have chosen to represent the data in such a way that the maximum velocity of the grains is 1/2, which is due to a total initial displacement of the perturbed edge grain of unity.

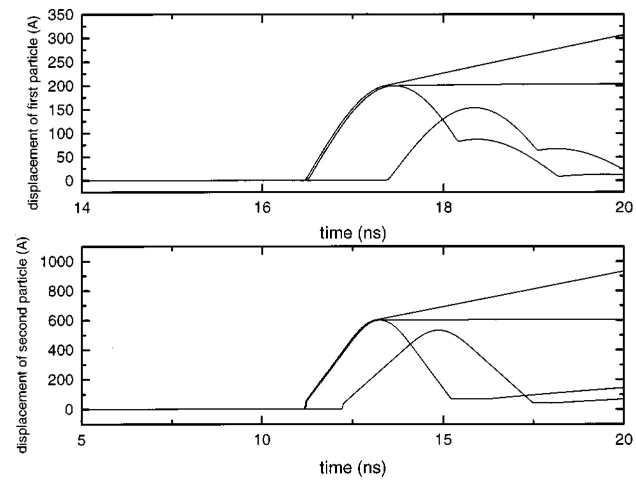

FIG. 3. Upper panel: plot of displacement of ferrofluid grains of diameter 20 nm at the surface of a ferrofluid for $v_{\mathrm{init}}=25.700$ m/s (for the case where displacement starts late), 33.441 m/s (for the case where displacement starts earlier but there is no escape), 33.443 m/s (barely escapes), and 33.700 m/s (escape). Lower panel: same as the upper panel for the ferrofluid grain second from the surface. The v_{init} values in the same order as above are 1500.0, 2304.0, 2305.16, and 2330.0 m/s.

the net effect of restitution is to introduce exponential decay of the amplitude of the propagating soliton without affecting its width.[10–12] Thus, given restitution, one can infer the magnitude of v_{init} required to eject a ferrofluid grain through the water–air interface.

Figure 3 describes the displacement of the *surface grain* as a function of time for various magnitudes of v_{init}. The data show that the surface grain is able to overcome the effect of surface tension for sufficiently strong v_{init} generated at the bottom of the ferrofluid. It is interesting to observe that when the surface particle is able to free itself from the ferrofluid, the second particle from the surface cannot do the same (Fig. 3). An impulse that is some 50 times larger is needed to successfully eject the second particle. This result can be understood by referring to Fig. 2, which shows that while the spatial extent of the soliton is about 5 ferrofluid grains, most of its kinetic energy is confined to a single ferrofluid grain at the center of the 5 grain package.

We next ask whether significantly smaller v_{init}s would suffice to eject ferrofluid grains of larger diameter. Consider a ferrofluid grain of radius R_0 and let us scale up its radius by s $(s>1)$ such that the new radius is $R=sR_0$. The initial mass m_0 becomes $m=s^3m_0$ and according to Eq. (1), $a=(s)^{0.5}a_0$. The magnetic force $F_m=s^2F_{m0}$, the gravitational force Ferrofluid grain$=s^3$ Ferrofluid grain$_0$ and the maximum surface force becomes $F_s=sF_{s0}$. Thus, the escape distance to be traveled to overcome the surface tension scales as $D_{\mathrm{esc}}=sD_{\mathrm{esc0}}$. Since escape of the ferrofluid grain from the liquid requires the input kinetic energy to equal the energy associated with surface tension (E_s), one can write, $mv_{\mathrm{init}}^2/2=E_s$. This equation implies that the new $v_{\mathrm{init\text{-}new}}=v_{\mathrm{init}}/(s)^{0.5}$. Our study confirms this result via direct particle dynamical calculations of impulse propagation through a chain. The results establish that increasing the diameter of the ferrofluid grains helps in reducing the magnitude of the initial impact without making a major sacrifice in the droplet size.

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