

Dynamics of particle suspensions in heterodyned triaxial magnetic fields

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Abstract

We have begun to explore the striking collective dynamics that can occur when a magnetic particle suspension is exposed to three orthogonal ac magnetic fields. The motivation for doing this requires some explanation. When a uniaxial magnetic field is applied to a magnetic particle suspension, the induced dipoles cause the formation of particle chains. We call this a positive dipolar interaction. If instead an audio frequency rotating magnetic field is applied in the plane normal to the uniaxial field, the suspended particles will form parallel sheets, due to the creation of negative dipole interactions. If both a uniaxial and a rotating field of equal amplitude are applied simultaneously, which we call a *balanced triaxial field*, the average dipolar interaction should vanish, but it doesn't. Why? Because each dipole moment has a component due to the applied field, and a component due to the fields of all the other dipoles. The former does indeed give a null average interaction, but the latter does not. This non-vanishing dipole field interaction is terribly complex: it cannot be crudely represented as a sum of pair interactions, even for three dipoles. Because of this the usual structures one expects of attractive interactions - spinodal structures, equilibrium clusterings, lattice formation - fail to emerge. Instead, a variety of particle networks and particle foams form, these having large magnetic susceptibilities. Unusual dynamical phenomena can be induced by setting two or more of the coil frequencies sufficiently close that the suspension can follow the beats. If this heterodyning is done during the gelation of the suspension resin, some surprising particle structures emerge, such as a cellular honeycomb. These structures have highly optimized physical properties. We will discuss both experimental work and molecular dynamics simulations of these phenomena.

1 Introduction

When a random suspension of magnetically soft particles is exposed to a uniaxial magnetic field, dipoles are induced that are closely aligned with the applied field.

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The particles will move under the influence of the dipolar interactions with nearby particles in such a fashion as to increase their dipole moments, forming complex chain-like structures that reduce the suspension magnetostatic energy. We call interactions in a uniaxial field *positive* dipolar interactions. These interactions are invariant to inversion of the field, since this merely inverts the sign of each dipole and the interactions depend on the square of the dipole moment. A *negative* dipolar interaction between two particles can in principal be created by inverting the field at only one of the dipoles, but this is impractical, and in any case it would not be possible to create negative dipole interactions between all particles with such an approach.

It is possible to create a time-average *negative* dipole interaction between particles by subjecting the suspension to a rapidly changing biaxial (e.g. rotating) magnetic field in a plane normal to the uniaxial field described above. The induced dipole moments create a net average attractive interaction in the plane of the field, resulting in the formation of complex sheet-like structures. [1] A simple first-order calculation [2] shows that the average interaction is just the opposite of a positive dipolar interaction, so that in a balanced triaxial field, where both the uniaxial and biaxial fields are applied simultaneously (and with all three field components having equal rms amplitudes), one might expect no interaction at all. Experiments show that this is not the case, and an exact, self-consistent point dipole calculation shows that the negative dipolar interactions created by a biaxial field are not quite equal and opposite to the positive dipolar interactions.

This lack of perfect symmetry between interactions in uniaxial and biaxial fields is due to the fact that the particles magnetize in the local field, not the bare applied field, which can be viewed as a sum of the macroscopic field and the microscopic field. The smooth macroscopic field is the sum of the applied field plus the Lorentz cavity field (provided demagnetizing fields due to the sample shape can be eliminated). The lumpy microscopic field is due to the nearby dipoles. Each dipole moment thus has parts due to the macroscopic and microscopic fields. It can be shown that in a balanced triaxial field the part of the moments due to the macroscopic field lead to zero average interaction, but the part due to the microscopic field lead to strong and complex many-body interactions. Complex because the interaction between three contacting particles cannot even be approximately described by the sum of the pair potential. Because of this, all of the usual things one might expect of an isotropic system with attractive interactions, such as the formation of periodic lattices, fail to occur. Instead, particle structures with molecular-like geometries are stable, and at equilibrium a suspension forms a particle foam. [3]

By the judicious selection of the triaxial field component amplitudes and frequencies a variety of complex particle suspension dynamics can be created, including vortex motion and particles sheets that seem to chaotically explore a variety of orientations. If the suspending fluid is a polymerizable resin, the suspension dynamics can lead to novel particle composites with optimized physical properties. If all three field frequencies are widely separated a particle gel forms. If two of the field component frequencies are sufficiently close

together that the particles can follow the beat frequency, then a striking oscillation occurs we call 2-d heterodyning, which leads to the formation of a honeycomb structure. If all three component frequencies are close, a complex collective dynamics occurs we call 3-d heterodyning, which leads to a particle foam. The rms field amplitudes can also be imbalanced to create structures with anisotropic magnetic properties. We consider both positive and negative uniaxial biases, which lead to composites with enhanced magnetic susceptibilities along one or two principal directions, respectively.

2 Experimental

2.1 Sample preparation

For the studies of suspension dynamics, 50 μm Ni powder from Fisher Scientific was suspended in silicone oil, with concentrations in the range of 0.1-7.0 vol.%. For encapsulated structures the particles were suspended in the polyester resin Castin' Craft™. After addition of the catalyst, these suspensions were degassed in a vacuum chamber for 3 minutes, and poured into 1 cm square polystyrene cuvettes. Each cuvette was filled to a height of 1 cm to insure that the demagnetizing fields are the same in all directions. The samples were then structured in the triaxial magnetic field until well past the gel point (25 minutes) and post-cured in an oven at 70°C for at least two hours.

2.2 Triaxial field generation

To create a uniform triaxial magnetic field we constructed three nested orthogonal Helmholtz coil pairs. Although all three fields are equivalent, it is useful to think of one component as the uniaxial field and the other two as forming a biaxial field. The uniaxial field either is run dc with a current source or is driven ac with a fixed capacitor in series to create a series resonance at 203.7 Hz with an impedance of ≈ 2.6 ohm. The biaxial field components are run ac and are connected to tunable, computer-controlled, series-parallel capacitor banks of our own design to create series LRC circuits with high quality factors and an resonant impedance of ≈ 2.2 ohm. Driving these circuits with an ATI model 1504 audio power amplifier with 200 W per channel it is possible to create magnetic induction fields as large as 500 G at frequencies up to ≈ 1500 Hz. [4]

3 Simulation method

We have reported athermal [1,5] and thermal [6] simulation studies of structure formation in uniaxial and biaxial field-structured composites, but these simulations were based on the fixed point dipole approximation (i.e. magnetization in the applied field), which would give zero interactions in a triaxial field. A new simulation was written to compute interactions in a triaxial field. This simulation is exact in the self-consistent point dipole approximation, the primary difference being the consideration of the microscopic field in the

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computation of the dipole moments. In addition, other aspects of the simulation were modified to create a robust, stable code.

In this Brownian dynamics simulation the particles are essentially hard spheres with induced dipolar interactions, Stokes friction against the solvent, and Brownian motion. The essentially hard spheres have an interaction force that increases as the sixth power of their *overlap*, specifically, $f \propto (1.03d - \Delta r)^6$ for $\Delta r < 1.03d$, where d is the particle diameter and Δr is the separation distance between the centers of masses of the particles. Note that "overlap" starts when there is actually a 3% gap between the particles. The hard force amplitude can be chosen in a number of ways, but we chose it such that two particles in a balanced triaxial field of unit rms component amplitudes would have a center of mass separation equal to d . Larger agglomerates, such as a chain of particles, will compress somewhat, an important issue to which we will return.

To compute the dipole interactions exactly in the point dipole approximation one must take into account that the field that magnetizes the dipoles is the local field, the applied field plus the field due to the other dipoles. In addition, the magnetization of a single isolated particle is actually affected by the field created by its magnetization, [7] limiting the susceptibility (MKS units) of a spherical particle to $\chi = 3\beta$, where $\beta = (\chi_p - \chi_l) / (3 + \chi_p + 2\chi_l)$ in terms of the intrinsic susceptibilities χ_p and χ_l of the materials of which the particle and liquid phases are composed. For magnetic particles β can be as large as 1; for magnetic holes, as small as -1/2. The moment of a particle of volume v can then be written in terms of the local field as $\mathbf{m} = 4\pi\beta v \mathbf{H}_{local}$, and the force on a dipole is $\mathbf{F} = \mu_0 \mathbf{m} \cdot (\nabla \mathbf{H}_{local})$. In a non-heterodyned triaxial field we have shown that it is only necessary to sum the force for fields applied along three orthogonal axes [3].

The computation of the local field is a subject about which there is a certain degree of confusion in the literature [8]. We use the method of Lorentz (not to be confused with the Lorentz approximation), which enables the errors in the local field to be made as small as desired. The simulation volume is a cube with cyclic boundary conditions.

The macroscopic field at each dipole is taken to be the applied field plus the Lorentz cavity field, which is exactly $\mathbf{M}/3$ for the spherical cavity we employ (see below), with $\mathbf{M}$ the sample magnetization. The demagnetizing field due to the overall shape of the simulation volume is zero because when we compute the field at each dipole the simulation volume is always taken to be centered on that dipole, which is another way of using the cyclic boundary conditions, in addition to particle reentry.

The microscopic field at each particle is computed by summing the dipole fields $\mathbf{H}_m = [3(\mathbf{m} \cdot \hat{\mathbf{r}})\hat{\mathbf{r}} - \mathbf{m}] / 4\pi r^3$ from all the particles within a spherical cavity with a diameter that in practice is not less than 10x the particle diameter. Here $\hat{\mathbf{r}}$ is the unit vector from the particle center of mass. These nearby dipoles are computed with an efficient neighbors algorithm used in our previous simulations. The sum of the microscopic field and the Lorentz cavity field is the total field from all the dipoles and their images. Errors can result from this approach if the cavity size is smaller than the characteristic internal scale of the structure. In

practice this method is sufficiently precise that when we calculate the sample magnetization by enlarging the spherical cavity to include the entire simulation volume, the magnetization changes by $\approx 1.0\%$. Finally, local update is used in the calculation of the dipole moments because we found that global update could result in oscillation instabilities.

One critical numerical issue arises because a system of soft magnetic particles is perilously close to spontaneous magnetization. We have shown above that the susceptibility of a typical soft magnetic particle is quite close to 3. A straightforward calculation shows that a long chain of contacting particles will magnetize spontaneously if the particle susceptibility is $6/\zeta(3) \approx 5$, where $\zeta(x)$ is the Riemann zeta function. If the chain is slightly compressed to force the particles to overlap, spontaneous magnetization will occur at a center of mass separation of $d[\zeta(3)/2]^{1/3} \approx 0.844d$, at which point the dipolar force is infinite. Because of the proximity of this force divergence it is easily possible to choose a pseudo-hard sphere potential that is insufficiently stiff to prevent an agglomerate from collapsing to a point. There are two ways around this problem. First, one can choose an extremely stiff potential. This is a poor solution because it leads to force gradients that are extremely large, necessitating unreasonably small time steps for the particle dynamics to remain stable. Second, one can simply limit the dipole force and dipole field from overlapping particles to that which would obtain if the particles were exactly in contact. This approach eliminates "black hole" formation and enables the use of a pseudo-hard sphere potential that does not generate terrific force gradients, permitting a long time step.

The final simulation issue is stability. Our code is stabilized by keeping track of the force gradients and limiting the time step in proportion to the inverse gradient. In practice, during a simulation cycle each particle is locally advanced through the entire time interval, but this time interval is subdivided into smaller time steps if the program determines that the force gradients are unacceptably large. This is effective in reducing execution time and allows the code to make 10,000 particle runs on a Macintosh G4 in a reasonable amount of time.

The simulation is run in dimensionless time units. The athermal equation of motion is obtained by balancing the dipolar force, against the hydrodynamic drag force $F_{\text{hydro}} = -6\pi\eta av$, where a is the particle radius, η is the liquid viscosity and v is the particle velocity. The many-body dipolar force cannot be written in closed form, but is of the form $F_{\text{dip}} = a^2\mu_r\mu_0H_0^2f_{\text{dip}}$, where $\mu_0 = 4\pi \times 10^{-7} \text{ Wb/(Am)}$ is the vacuum permeability, μ_r is the relative permeability of the liquid phase, H_0^2 is the mean square field amplitude and f_{dip} is a dimensionless force. This results in a dimensionless equation of the form $\Delta u = \Delta f(r, \theta)$, where the dimensionless length $u = r/2a$ and the dimensionless time is $s = t/\tau$ with $\tau = 12\pi\eta/\mu_0H_0^2$. For a nonmagnetic suspending liquid with a viscosity η_0 of 1 cp, this characteristic time τ is 1 ms with an applied field of $H_0 = 5.5 \times 10^3 \text{ A/m}$ (69 Oe). The simulation data we generate are for structures that have evolved for 100 dimensionless time units or less.

The magnitude of the Brownian force is set by the parameter $\lambda = \pi\kappa^3\mu_r\mu_0H_0^2/k_B T$. In most of the thermal anneals reported here, we start with

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$\lambda=2.67$, anneal for 20 dimensionless time units, then linearly ramp temperature (i.e. $1/\lambda$) down to zero in 5 dimensionless time units. In some of the anneals we start at the same temperature but linearly ramp to zero over 25 time units. For 10,000 particles this takes a little less than 3 days on a 1 GHz Macintosh G4. Heterodyned simulations were athermal and took 12 days to achieve 100 dimensionless time units.

4 Dipolar interactions in heterodyned fields

Heterodyning of the field components leads to interesting suspension dynamics, and some discussion of this effect is helpful. We start by addressing the issue of dipole interactions in time-dependent fields. A particle of moment $\mathbf{m}_i$ is magnetized by the local field $\mathbf{H}_{loc} = \mathbf{H}_0 + \sum_{k \neq i} \mathbf{H}_{i,k}$, where

$\mathbf{H}_{i,k} = [3(\mathbf{m}_k \cdot \hat{\mathbf{r}}_{ik})\hat{\mathbf{r}}_{ik} - \mathbf{m}_k]/4\pi r_{ik}^3$ is the field at the i -th particle due to the k -th dipole. The force on this dipole is $\mathbf{F}_i = \mu_0 \mathbf{m}_i \cdot (\nabla \mathbf{H}_{local})$, which can thus be written as $\mathbf{F}_i = \mu_0 \mathbf{m}_i \cdot \sum_{k \neq i} \nabla \mathbf{H}_{ik}(\mathbf{m}_k, \mathbf{r}_{ik})$. Each contribution f_{ik} to the force is a

linear function of the moments, so $\mathbf{F}_i = \sum_{k \neq i} f(\mathbf{m}_i, \mathbf{m}_k)$ where

$f(\lambda \mathbf{m}_i, \gamma \mathbf{m}_k) = \lambda \gamma f(\mathbf{m}_i, \mathbf{m}_k)$. To understand heterodyning we need only consider the contribution to the total force from any one pair of particles in a particle agglomerate, which should not be taken to mean that interactions in the agglomerate can be described by summing a pair potential!

4.1 Coherent and incoherent interactions

For simplicity we start with a rotating field in the x - y plane, $\mathbf{H}_0 = H_0 [\cos(\omega t)\hat{\mathbf{x}} + \sin(\omega t)\hat{\mathbf{y}}]$, and ask what the force of interaction is when averaged over one cycle of the field. This field is at an angle $\phi = \omega t$ to the x axis, so the instantaneous field is $\mathbf{H}_0 = H_0 (\cos \phi \hat{\mathbf{x}} + \sin \phi \hat{\mathbf{y}})$. The self-consistent moment of the k -th dipole is $\mathbf{m}_k = \mathbf{m}_{x,k} \cos \phi + \mathbf{m}_{y,k} \sin \phi$, where $\mathbf{m}_{x,k}$ is the moment with applied field along $\hat{\mathbf{x}}$. The interaction force is $f_{i,k}(\phi) = f(\mathbf{m}_{x,i} \cos \phi \hat{\mathbf{x}} + \mathbf{m}_{y,i} \sin \phi \hat{\mathbf{y}}, \mathbf{m}_{x,k} \cos \phi + \mathbf{m}_{y,k} \sin \phi)$, or

$$f_{k,i}(\phi) = f_{k,i}(0) \cos^2 \phi + f_{k,i}(\pi/2) \sin^2 \phi + [f(\mathbf{m}_{x,i}, \mathbf{m}_{y,k}) + f(\mathbf{m}_{y,i}, \mathbf{m}_{x,k})] \cos \phi \sin \phi \quad (1)$$

To this force add the value when the applied field is at 90° to ϕ , $f_{k,i}(\phi \pm \pi/2)$. The cross terms cancel, with the result $f_{k,i}(\phi) + f_{k,i}(\phi \pm \pi/2) = f_{k,i}(0) + f_{k,i}(\pi/2)$. The interaction force between a pair of dipoles in an aggregate of dipoles, when summed for two orthogonal fields *applied at separate times*, is thus independent of the angle ϕ . The average interaction force in a rotating field is a simple

average of the forces in orthogonal fields,

$$\frac{1}{2}f_{k,j}(0) + \frac{1}{2}f_{k,i}(\pi/2) = \frac{1}{2}f(\mathbf{m}_{x,i}, \mathbf{m}_{x,k}) + \frac{1}{2}f(\mathbf{m}_{y,j}, \mathbf{m}_{y,k}). \quad (2)$$

so the dipoles are interacting *incoherently* when the x - and y - field components are in quadrature phase. In other words, the dipole moments when the applied field is in the x direction do not interact with the dipole moments when the applied field is in the y direction.

Compare this result to applying the field components in phase, to create an ac field $\mathbf{H}_0 = H_0 [\sin(\omega t)\hat{\mathbf{x}} + \sin(\omega t)\hat{\mathbf{y}}]$ at 45° to the x -axis. In this case the cross terms

$$[f(\mathbf{m}_{x,k}, \mathbf{m}_{y,k}) + f(\mathbf{m}_{y,k}, \mathbf{m}_{x,k})] \cos(\pi/4) \sin(\pi/4) \quad (3)$$

must be added to Eq. 2 to obtain the correct force. This is a *coherent* interaction between dipoles. In heterodyned fields there is an oscillation between coherent and incoherent interactions, which give rise to the unusual suspension dynamics.

4.2 Two-dimensional heterodyning

To create heterodyning in a biaxial field one adjusts the component frequencies to be sufficiently close that the beat frequency $\Delta\omega$ is slow enough for the particle suspension to follow it. The field can then be written as $\mathbf{H}_0 = H_0 \{\sin[(\omega + \Delta\omega/2)t]\hat{\mathbf{x}} + \sin[(\omega - \Delta\omega/2)t]\hat{\mathbf{y}}\}$ since the relative phase of the two sinusoids is no longer important. Under the conditions where $\Delta\omega \ll \omega$ the sinusoids will oscillate many times before the "relative phase" term $\alpha = \Delta\omega t$ changes, so that over a small time interval the field can be thought of as $\mathbf{H}_0 = H_0 [\sin(\omega t + \alpha)\hat{\mathbf{x}} + \sin(\omega t)\hat{\mathbf{y}}]$ within an arbitrary overall phase.

Writing the phase as $\alpha = n\pi/4$, a plot of H_x versus H_y reveals the following sequence: $n=0$ gives a linear (ac) field at 45° ; $n=1$ gives a clockwise elliptical field whose major axis is at 45° to the x axis; $n=2$ gives a clockwise rotating field; $n=3$ gives a clockwise elliptical field whose major axis is at 135° ; $n=4$ gives a linear (ac) field at 135° ; $n=5$ gives a counterclockwise elliptical field whose major axis is at 135° ; $n=6$ gives a counterclockwise rotating field; $n=7$ gives a counterclockwise elliptical field whose major axis is at 45° ; and $n=8$ restarts the sequence. Intermediate values of n always correspond to elliptical fields at either 45° or 135° to the x axis. When this heterodyned biaxial field is combined with an orthogonal uniaxial field at a significantly different frequency, an oscillation between a triaxial field and a biaxial field occurs. This is apparent in the simulated dynamics in Fig. 1, an alternation between particle sheets and foams.

A straightforward generalization of Eq. 1 to arbitrary field component phasing gives the contribution of the field produced by dipole k to the force on dipole i at the time given by $\phi = \omega t$,

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$$f_{k,i}(\phi) = f_{k,i}(0) \sin^2(\phi + \alpha) + f_{k,i}(\pi/2) \sin^2 \phi + [f(\mathbf{m}_{x,i}, \mathbf{m}_{y,k}) + f(\mathbf{m}_{y,i}, \mathbf{m}_{x,k})] \sin(\phi + \alpha) \sin \phi \quad (4)$$

To obtain the average interaction during a single beat of the "carrier" frequency ω we compute the short-time average of Eq. 4,

$$f_{k,i}(\alpha) = \frac{1}{2} f_{k,i}(0) + \frac{1}{2} f_{k,i}(\pi/2) + [f(\mathbf{m}_{x,i}, \mathbf{m}_{y,k}) + f(\mathbf{m}_{y,i}, \mathbf{m}_{x,k})] \cos(\Delta\omega t). \quad (5)$$

This shows that the incoherent force has added to it a coherent cross term that is modulated at the heterodyning beat. In computer simulations of heterodyning one can use a force averaged over the carrier cycle, reducing the computation time by an enormous amount, and eliminating ω as a variable. This is correct to do when structural evolution times are slow compared to $1/\omega$ and fast compared to $1/\Delta\omega$. During gelation structural evolution times ultimately become slow compared to $1/\Delta\omega$, resulting in the formation of a honeycomb structure.

A time average of the heterodyned interaction force Eq. 5 over a single cycle of the beat frequency recovers the incoherent force in Eq. 2. Thus if the beat is rapid compared to structural organization times the evolution of structure is unaffected by the beat, except that memory effects persist.

4.3 Three-dimensional heterodyning

In three dimensional heterodyning all three fields beat at a low frequency. It is not easy to envision the dynamically changing Lissajou loop in this case. To understand 3-d heterodyning it is useful to write Eq. 5 in the form (dropping the subscripts i, k) $f(t) = \frac{1}{2} f_{xx} + \frac{1}{2} f_{yy} + f_{xy} \cos(\Delta\omega_{xy} t)$. In this simplified notation f_{xx} refers to the force between dipoles in a uniaxial field of amplitude H_0 applied along the x axis, whereas $f_{xy} = f_{yx}$ is the cross term. To obtain the general case define the matrix $\mathbf{F}$, such that $\mathbf{F}_{xy} = f_{xy}$. Defining the symmetric heterodyning matrix

$$\Omega = \begin{bmatrix} 1 & \cos(\Delta\omega_{xy} t) & \cos(\Delta\omega_{xz} t) \\ \cos(\Delta\omega_{xy} t) & 1 & \cos(\Delta\omega_{yz} t) \\ \cos(\Delta\omega_{xz} t) & \cos(\Delta\omega_{yz} t) & 1 \end{bmatrix}. \quad (6)$$

gives the compact expression $f = \frac{1}{2} \Omega \mathbf{F}$ for the force averaged over the carrier frequency. One beat frequency is the sum of the other two, so there are only two independent beat frequencies. If there is a definite phase relation between the fields, it is simple to introduce these phases into the heterodyning matrix. Eq. 6 is the force used in our heterodyning simulations.

Three dimensional heterodyning creates a particle dynamics wherein sheets appear in an apparently unpredictable manner normal to the four body diagonals

of a cube whose faces are normal to the field components, especially when the beat frequencies are related to each other by a ratio of large integers.

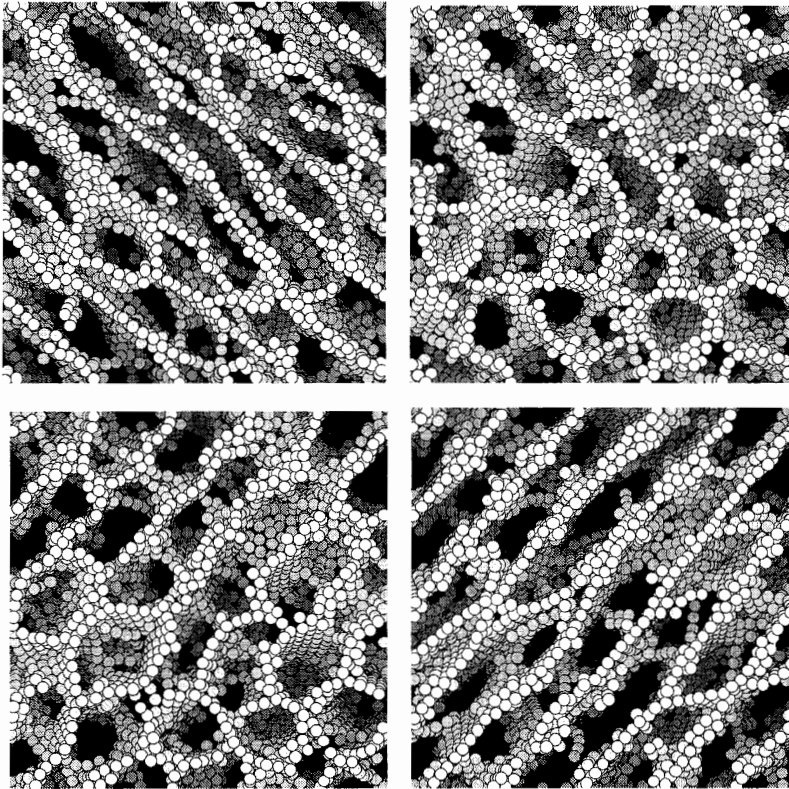


Figure 1: Series of structures during 2-d heterodyning of a triaxial field. View is along the uniaxial field, i.e. normal to the heterodyned biaxial field. The periodic formation of sheets occurs when the heterodyned field in the x-y plane becomes linear. This linear ac field combines the uniaxial field to create a pure biaxial field, and sheets form in this field plane. During gelation this produces a honeycomb structure.

4.4 Gelation

In a gelling liquid the viscosity diverges to infinity at the gel point. Because of this divergence, a heterodyned particle suspension will no longer be able to couple to the beat at some time before gelation, which has interesting and important effects on structure. According to dynamical scaling theory [9] this viscosity divergence is $\eta \approx |t - t_{gel}|^{-4/3}$. One approach to gelation is to incorporate this result into a heterodyning simulation. It is more efficient to chirp the heterodyne frequencies. For simulations of 100 dimensionless time units we used



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$\Delta\omega = \Delta\omega_0/(1-t/105)$ as the chirp function, with the initial beat frequencies close to one reciprocal dimensionless time unit.

5 Conclusions

We have shown that it is possible to use triaxial magnetic fields to create complex particle interactions. These interactions are especially interesting when heterodyning of the field components is employed, to produce slow oscillations between coherent and coherent particle interactions. Although the suspension dynamics itself is interesting, polymerization of the continuous phase during these dynamics produces composite materials with highly optimized properties, so the application of triaxial fields is a method of producing improved materials.

Acknowledgments: Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy under contract DE-AC04-94AL8500. This work supported by the Division of Materials Sciences, Office of Basic Energy Sciences, U.S. Department of Energy (DOE).

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