Novel Convective Instabilities in a Magnetic Fluid

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(Received 10 December 1998)

A field induced novel instability was observed in a horizontal magnetic fluid layer placed in a uniform vertical magnetic field with a focused laser beam parallel to the field passing through the layer. The unstable diffraction patterns include triangles and other polygons. We found that the critical field for the onset of this instability increases with increasing the thickness of the layer. These results are consistent with our linear stability analysis. A new bifurcation was also observed when an additional horizontal field was applied. [S0031-9007(99)09113-9]

PACS numbers: 83.10.Ji, 47.27.Te, 66.10.Cb, 82.40.Bj

Nonlinear dynamics outside equilibrium is one of the intriguing and ubiquitous phenomena in nature. Exploring new mechanisms for driving forces and understanding relations between structures and dynamics will not only elucidate the underlying physics but also lead to new applications. In most research on instabilities, the driving forces involve gravity, surface tension, and ponderomotive force [1,2]. In this Letter, we report the observation of a novel instability in a horizontal magnetic fluid layer with a focused laser beam perpendicularly passing through the layer in the presence of a homogeneous vertical magnetic field. We found that the diffraction pattern formed by the laser beam remained stable when the field was less than a critical value, whereas it became unstable when the field was larger than the threshold. This instability is driven by a force that was rarely studied before—the magnetic Kelvin body force that originates from the interaction of the magnetic moment of the fluid with the internal field gradient induced by temperature and concentration inhomogeneities, *even though the external field is uniform in space.* This force can be easily adjusted, by changing the external field and the laser power, to control heat and mass transfers in the system. Studies of this novel instability should lead to not only new insights in fundamental research on nonequilibrium phenomena but also practical applications such as field-controlled heat transfer devices and liquid optical devices.

The magnetic fluid [3] used in our experiment consists of magnetite particles (volume fraction 6%) suspended in kerosene. The mean diameter of each particle is 9 nm and each particle is coated with a nonmagnetic surfactant layer of 2 nm in thickness to prevent agglomeration. In Fig. 1 we show the schematic diagram for the experimental setup. The fluid sample with a thickness of 100 μm is sandwiched between two horizontal parallel glass plates with a dimension of 2×2 cm^2. A 10 mW He-Ne laser beam with a wavelength of 632.8 nm is focused normally on the sample from below by a lens. The radius of the beam is 0.6 mm and the focal length of the lens is 2 cm. The diffraction patterns at far field above the sample are captured by a charge-coupled device camera and are digitized by a computer. The details about the experimental setup and the sample can be found elsewhere [4].

In the absence of magnetic fields, multiple concentric rings are observed after the laser is switched on and they reach a steady state in a time scale of seconds. The stable rings are illustrated in Fig. 2(a). When a laser beam with a Gaussian profile is focused on the sample cell, the fluid's strong absorption of light (absorption coefficient 550/cm) leads to a temperature distribution that decreases monotonically in the radial direction about the beam. The

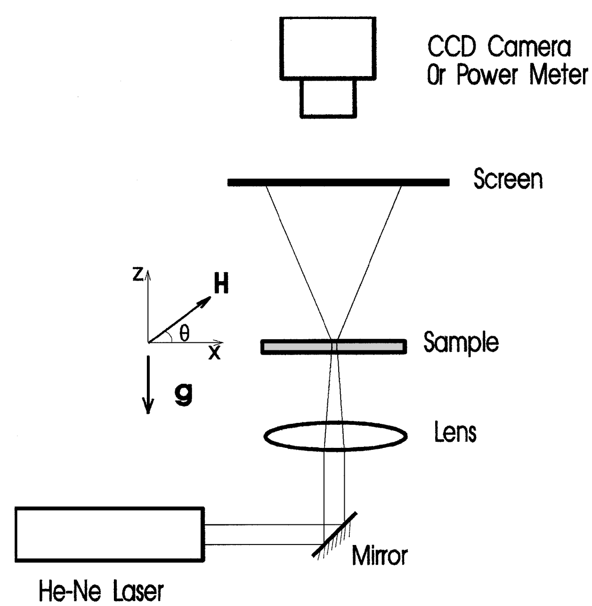

FIG. 1. Schematic of experimental setup. Both the acceleration of gravity and the laser beam are along the z direction. The direction along the in-plane component of the applied field is defined as x axis. To show the sample and field orientations, two pairs of Helmholtz coils are omitted.

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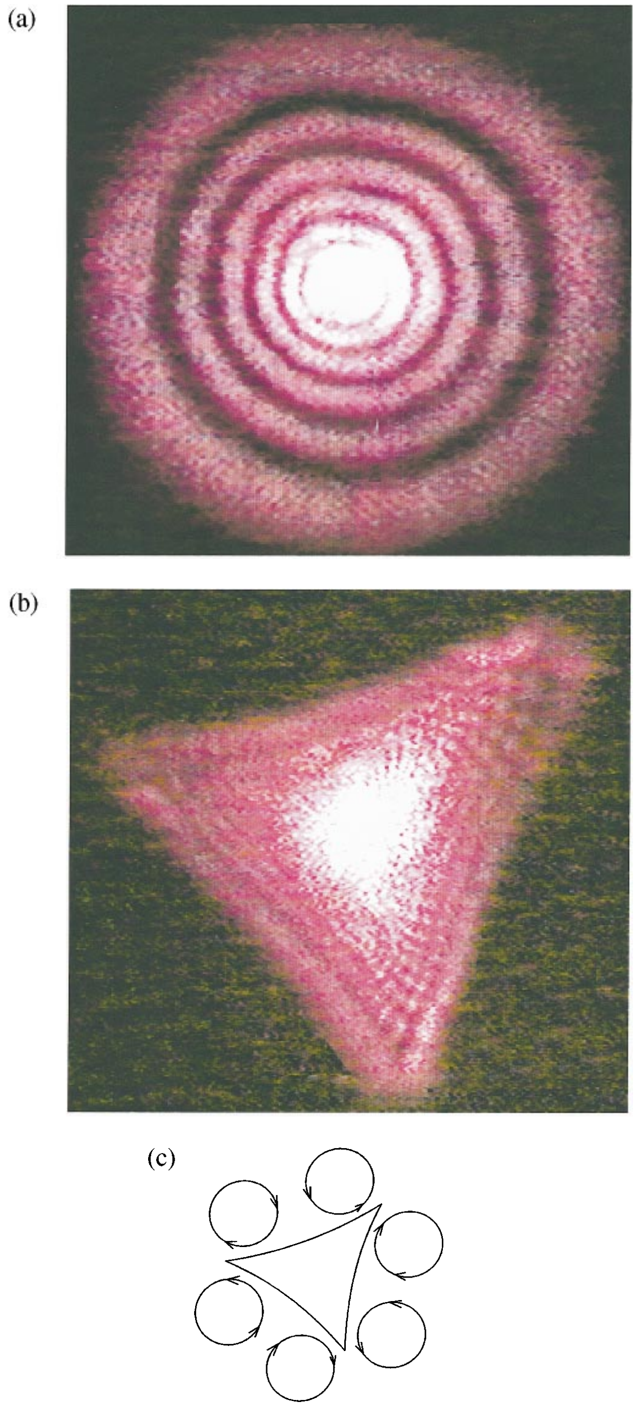

FIG. 2. The far-field diffraction pattern shows the onset of instability unambiguously. (a) (color) In zero field, the concentric rings form when a laser beam is focused to the sample layer. The symmetry of the rings reflects the axisymmetry of the laser beam. (b) (color) When the applied field exceeds a threshold, H_c, the instability starts then the circular rings are replaced by "polygons." Shown here is a triangle, one of the observed unstable patterns. (c) The triangular shape in (b) originates from six convective rolls with their axes parallel to the optical axis.

effect of the focusing lens is to increase the intensity of the laser from 10^5 W/m^2 to 10^8 W/m^2 and hence to create a significant temperature gradient. The radius of the focused beam is 6.7 μm. Our numerical calculation [5] shows that the temperature difference is about 15 K between the beam axis and the beam edge. However, the temperature continues to decrease outside the beam and levels off at several beam widths; the corresponding temperature difference is about 40 K. This thermal gradient induces a particle-concentration gradient via the so-called Soret effect [6]. The positive Soret constant for our fluid [5,7], $S_T \sim 10^{-1}$ K^{-1}, much larger than many binary liquids [8], leads to a large positive concentration gradient of particles. These radial gradients of concentration and temperature yield the radial distribution in the index of refraction of the fluid, producing the observed circular rings which reflect the axisymmetry of the power distribution.

In the presence of a magnetic field, the magnetic fluid experiences a Kelvin body force per unit volume $\mathbf{f}_m = \mu_0(\mathbf{M} \cdot \nabla)\mathbf{H}'$, which arises from the interaction between the local magnetic field $\mathbf{H}'$ within the fluid and the magnetic moments of the particles characterized by the magnetization $\mathbf{M}$. Here, μ_0 is the permeability of free space. This force tends to move the fluid to higher field regions. For small magnetic fields, $\mathbf{M} = \chi(T, C)\mathbf{H}'$, where χ is the magnetic susceptibility of the fluid following Curie's law, $\chi \propto C/T$. In the presence of a uniform external vertical field $\mathbf{H}$, the internal magnetic field in the layer of the fluid has the form $\mathbf{H}' = \mathbf{H}/(1 + \chi)$. Since $\nabla \times \mathbf{H}' = 0$, the Kelvin body force follows as

$$\mathbf{f}_m = \frac{1}{2}\,\mu_0\chi\nabla H'^2 = \frac{\mu_0\chi^2H^2}{(1 + \chi)^3}\left(\frac{\nabla T}{T} - \frac{\nabla C}{C}\right), \quad (1)$$

where H is the magnitude of $\mathbf{H}$ and H' the magnitude of $\mathbf{H}'$. Thus both temperature and concentration gradients can make this force spatially nonuniform even if the external field is uniform. It shows that the inward radial thermal gradient and the outward radial concentration gradient render $\mathbf{f}_m$ centripetal in the radial direction. Because of these radial gradients in temperature and concentration, the mass density of the fluid increases in the radial direction. Since the magnitude of this centripetal force also increases in this direction, the fluid is potentially unstable in the sample plane. When this destabilizing force is strong enough to overcome the stabilizing dissipative effects of viscosity, thermal conduction, and particle diffusion, the fluid becomes unstable. Therefore, there exists a critical value H_c for the applied field H above which instability sets in.

By placing a pair of Helmholtz coils along the vertical direction (parallel to the laser beam), we can apply an adjustable dc magnetic field on the fluid. We observed the existence of a threshold ($H_c \sim 22$ Oe) for the field [9]. When $H < H_c$, the fluid is stable and the circular rings remain steady. However, these rings become unstable for $H > H_c$, indicating the onset of instability. We observed different diffraction patterns including "triangle" like shapes as shown in Fig. 2(b), "tetragon," and "pentagon." These patterns switch among different shapes alternately. The switching rate increases with increasing the field but no qualitative changes were observed within our field range.

To analyze this instability, we consider a two-dimensional fluid flow in the sample plane confined between two laterally unbounded parallel lines with a temperature difference ΔT across them. The fluid flow is governed by

$$\rho(\partial/\partial t + \mathbf{v} \cdot \nabla)\mathbf{v} = -\nabla P + \rho \nu \nabla^2 \mathbf{v} + \mathbf{f}_m , \quad (2)$$

$$(\partial/\partial t + \mathbf{v} \cdot \nabla)T = D_T \nabla^2 T , \quad (3)$$

$$(\partial/\partial t + \mathbf{v} \cdot \nabla)C = D_C[\nabla^2 C + S_T C(1 - C)\nabla^2 T], \quad (4)$$

$$\nabla \cdot \mathbf{v} = 0 , \quad (5)$$

where ν is the kinematic viscosity, D_T the thermal diffusivity, and D_C the concentration diffusivity. Our linear stability analysis yields the criterion for the onset of instability,

$$K > K_c = \frac{27\pi^4 Le}{4[(3 + \Psi)(1 + \Psi)Le + (2 + \Psi)\Psi]} , \quad (6)$$

where $\Psi = S_T(1 - C_0)T_0$ is the separation ratio, $Le = D_C/D_T$ the Lewis number, and

$$K = \frac{\mu_0 \chi_0^2 H^2 \Delta T^2 d^2}{\rho_0 T_0^2 (1 + \chi_0)^2 \nu D_T} \quad (7)$$

the Kelvin number. Here, the subscript 0 represents the reference point in the middle between the two lines and d the width between them. An estimation of the critical field H_c for H yields a value of the same order as observed, confirming our qualitative analysis.

To interpret the relation between the observed diffraction patterns and the configurations of the convective rolls, we notice that the laterally unbounded direction in the considered ideal system corresponds to the azimuthal direction within the sample plane with respect to the beam axis and the higher temperature side corresponds to the center of the beam. Therefore, the two-dimensional convective rolls are distributed along the azimuthal direction of the beam. The axes of these rolls are parallel to the beam axis. The number of the rolls must be even because of periodicity and axisymmetry. In a cylindrical shell containing a magnetic fluid with imposed radial magnetic and temperature gradients, a stability analysis [10] shows that convective states with six, eight, and ten rolls are the easiest states to excite. When $H > H_c$, the observed triangle diffraction pattern reflects a six-roll state as shown in Fig. 2(c). Any two adjacent rolls with outward radial flow between them form a corner and the six rolls form the triangle. Accordingly, an eight-roll state gives rise to a tetragon and a ten-roll state a pentagon. All these patterns have been observed in our experiments.

The radial temperature gradient in the fluid outside the laser beam is provided by the heat produced within the laser beam due to the strong light absorption of the particles in the fluid. So it is plausible to assume that this heat source maintains the imposed temperature difference in the two-dimensional model. Our calculation shows that the vertically averaged total heat per unit length is inversely proportional to the thickness of the sample layer L. Thus, the imposed temperature difference should also be inversely proportional to the thickness, i.e., $\Delta T \propto 1/L$, approximately. From Eqs. (6) and (7) we can conclude that the critical field is approximately proportional to the thickness, i.e., $H_c \propto L$ that fairly agrees with our observation as shown in Fig. 3(a). Contrary to the familiar magnetic Rayleigh-Bénard convection [11,12] and the magnetically controlled convection in nonconducting paramagnetic fluids [13,14], where critical values decrease with increasing the thickness of fluid layers, the critical field for the onset of instability in this system increases with the thickness. For a given L, Eqs. (6) and (7) imply that H_c should decrease with increasing the input laser power, which is also consistent with our experiments [see Fig. 3(b)].

By placing another pair of Helmholtz coils along the horizontal direction, we add a horizontal field H_x in addition to the vertical field H_z. Our observation shows that the instability is mainly controlled by H_z. However, we found the existence of another critical value H_c' for $H_x (H_c' \sim 10$ Oe). When $H_x < H_c'$, the diffraction patterns remained almost the same as without H_x. For $H_x > H_c'$, oscillations around H_x were observed. Figure 4 illustrates the

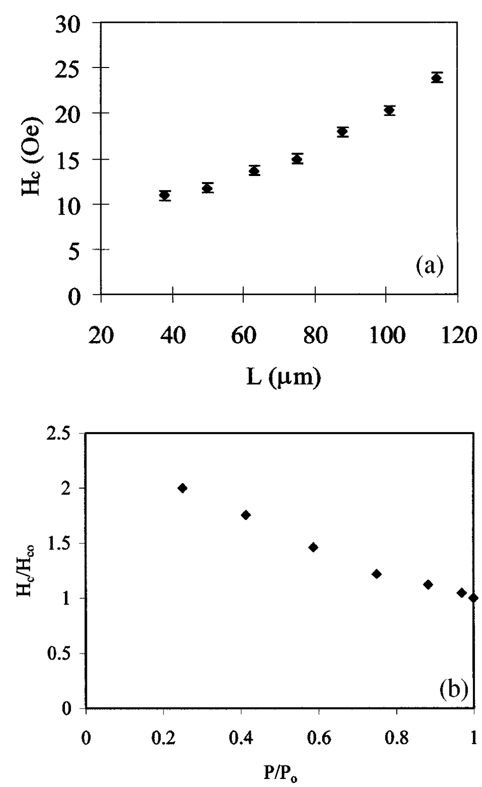

FIG. 3. (a) The critical field, H_c, increases with increasing sample thickness, L, for a given input laser power P; (b) H_c decreases with increasing P, for a given L. H_{c0} is the critical field for the input power $P_0 = 7$ mW.

(a)

(b)

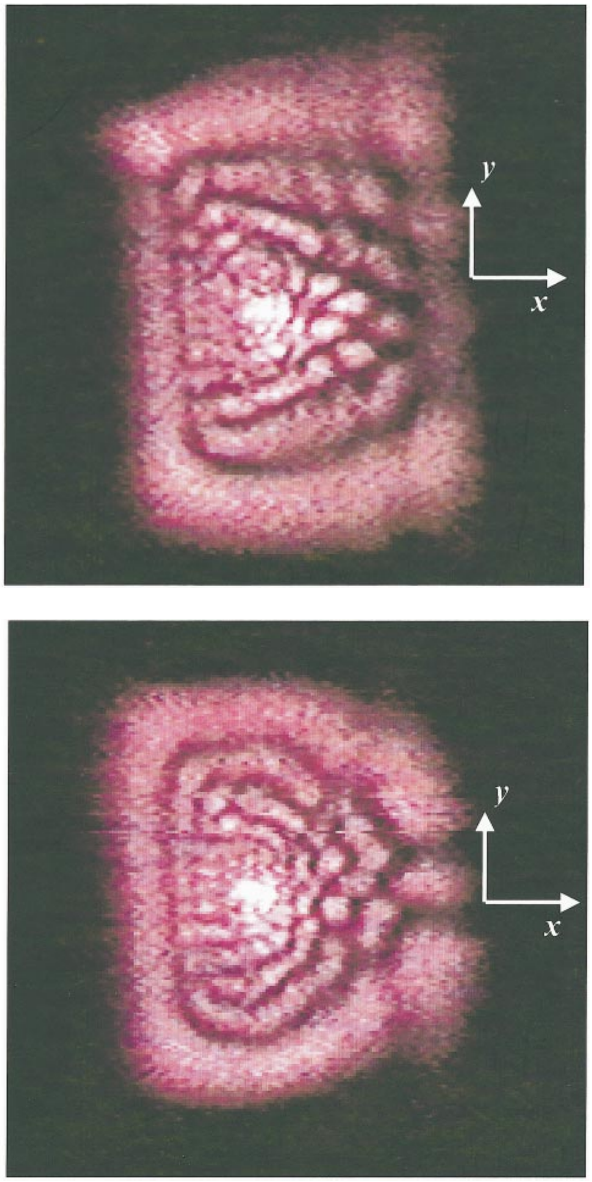

FIG. 4(color). When the horizontal component of the field, H_x, is larger than a certain value, oscillation of the diffraction pattern about the x axis occurs. The right side of the pattern first expands then shrinks or swings back and forth around the x axis.

instantaneous shapes of the pattern at times of half a period apart. The diffraction pattern oscillates in the sample plane (expands and shrinks in the direction perpendicular to the horizontal field). These results show that the new instability is induced by the horizontal field and therefore the interaction between H_x and the magnetic moment of the particles should be responsible for this new bifurcation. As the magnetic moment tends to align with the applied field, the horizontal field yields a nonzero horizontal moment, which breaks the axisymmetry of the system. Since any convective flow in the sample plane tends to turn the horizontal moment away from the horizontal field, this tendency in turn should yield an effect on the flow. For a small H_x, this effect is small and the diffraction patterns should remain almost the same as without H_x. However, a large H_x yields a large effect that could be strong enough to alter these patterns as observed. Its study could reveal the insights about the interactions between convective flows and magnetic fields.

In conclusion, a new convective instability was observed in a magnetic fluid. The corresponding mechanism is attributed to the interaction between the magnetization of the fluid and the magnetic field gradient originated from the inhomogeneity of both temperature and particle concentration. In the presence of the second field that breaks the axisymmetry of the system, an additional bifurcation was found, indicating the importance of symmetry breaking. The obtained results suggest that the heat and particle transfers in colloidal fluids could be monitored and controlled by an external field. Because the applied field also modulates the index of refraction, the results from this work demonstrate that the Kelvin force could also be used effectively in field-controlled liquid optical devices as well.

We thank Professor Carl Franck and Professor Albert Libchaber for useful discussions. We appreciate Dr. R. E. Rosensweig for providing the sample used in this experiment. One of us (W. L.) acknowledges support from the U.S. National Science Foundation.

[1] See, for example, S. Chandrasekhar, *Hydrodynamic and Hydromagnetic Stability* (Clarendon Press, Oxford, 1961).
[2] For a review, see M. C. Cross and P. C. Hohenberg, Rev. Mod. Phys. **65**, 851 (1993), and references therein.
[3] R. E. Rosensweig, *Ferrohydrodynamics* (Cambridge University Press, Cambridge, England, 1985).
[4] T. Du, S. Yuan, and W. Luo, Appl. Phys. Lett. **65**, 1844 (1994); T. Du and W. Luo, Mod. Phys. Lett. B **9**, 1643 (1995).
[5] T. Du and W. Luo, Appl. Phys. Lett. **72**, 272 (1998).
[6] C. Ludwig, Sitz.ber., Acad. Wiss. Wien **20**, 539 (1856); C. Soret, Arch. Sci. Phys. Nat. Geneva **2**, 48 (1879).
[7] N. V. Tabiryan and W. Luo, Phys. Rev. E **57**, 4431 (1998).
[8] P. Kolodner, H. Williams, and C. Moe, J. Chem. Phys. **88**, 6512 (1988).
[9] In *Proceedings of the Annual March Meeting of the American Physical Society, Kansas City* [Bull. Am. Phys. Soc. **42**, 775 (1997)].
[10] A. Zebib, J. Fluid Mech. **321**, 121 (1996).
[11] B. A. Finlayson, J. Fluid Mech. **40**, 753 (1970).
[12] J. Huang, B. F. Edwards, and D. D. Gray, Phys. Fluids **9**, 1819 (1997).
[13] D. Braithwaite, E. Beaugnon, and R. Tournier, Nature (London) **354**, 134 (1991).
[14] J. Huang, B. F. Edwards, and D. D. Gray, Phys. Rev. E **57**, R29 (1998).