Ferromagnetic resonance in periodic particle arrays

S. Jung, B. Watkins, L. DeLong, J. B. Ketterson and V. Chandrasekhar

Department of Physics and Astronomy, Northwestern University, Evanston, IL 60208, USA
Department of Physics and Astronomy, University of Kentucky, KY 40508, USA

We report measurements of the ferromagnetic resonance (FMR) spectra of arrays of submicron size periodic particle arrays of permalloy produced by electron-beam lithography. In contrast to plane ferromagnetic films, the spectra of the arrays show a number of additional resonance peaks, whose position depends strongly on the orientation of the external magnetic field and the interparticle interaction. Time-dependent micromagnetic simulations of the ac response show that these peaks are associated with coupled exchange and dipolar spin wave modes.

The physics of small ferromagnetic particles has become of increasing interest in recent years, driven to a great extent by their potential applications in high-density digital storage. As the size of the particles decreases, it is important to have access to experimental tools that can probe magnetic properties on the scale of nanometers. Conventional magnetic probes such as magnetometry are not sensitive enough to probe the properties of individual nanometer-scale particles. Recently developed tools such as magnetic force microscopy (MFM) can indeed image nanometer scale features, but their results are not always easy to interpret in terms of the intrinsic parameters of the ferromagnet. From this perspective, ferromagnetic resonance (FMR) is a powerful tool to probe the fundamental magnetic properties of ferromagnetic particles. As has been known for many years, FMR spectra are sensitive to the detailed geometry of the sample, and the FMR resonance shape depends on the intrinsic dissipation mechanisms in the ferromagnet. In addition, information about magnetic excitations such as spin waves can also be obtained. In spite of this, FMR has not been used extensively to probe the properties of nanometer scale ferromagnets. It is only recently that techniques such as magnetic resonance force microscopy (MRFM) have been applied to small magnetic structures, but such investigations are still in their infancy.

Consider then a ferromagnetic particle in a uniform external magnetic field \( H_0 \). Its magnetic moment \( M \) will evolve according to the Landau-Lifschitz equation:

\[
\frac{dM}{dt} = -\gamma M \times H_{\text{eff}} - \frac{\gamma \alpha}{M_s} M \times (M \times H_{\text{eff}})
\]

where \( \dot{M} \) is the rate of magnetic moment change, \( M \) is the magnetization, \( \gamma = ge/2mc \) is the gyromagnetic ratio (\( g \) being the Landé factor, and \( m \) the mass of the electron), \( H_{\text{eff}} \) is the effective local field, \( M_s \) is the saturation magnetization, and \( \alpha \) is the damping constant. This causes a precession of the moment about the direction of the field at a frequency \( \omega_0 \). For an ellipsoidal ferromagnetic body (with a uniform magnetization), \( \omega_0 \) is given by the Kittel equation:

\[
\omega_0^2 = \frac{\gamma^2}{M_s} \left[ H_0 + (N_x - N_z)M \right] \left[ H_0 + (N_y - N_z)M \right]
\]

where the \( N_x, N_y, \) and \( N_z \) are the demagnetization factors along the \( x, y \) and \( z \) directions which satisfy the relation \( N_x + N_y + N_z = 4\pi \), and the \( z \) direction is defined by the orientation of the external field \( H_0 \) and the magnetization \( M \). An ac magnetic field \( H_{ac} \) at this resonant frequency applied perpendicular to \( H_0 \) will couple to a uniform precession of \( M \) about the direction of \( H_0 \), resulting in absorption of energy from the ac field.

In addition to the uniform precession mode, one may also have non-uniform, or spin-wave, modes of precession. Kittel considered the case where the exchange interaction provided the dominant correction to the mode frequency. In his model, additional resonance peaks would be observed at frequencies \( \omega_p = Dk_p^2 + \omega_0 \), where \( D \) is a constant dependent on the exchange interaction between neighboring spins, and \( k_p \) is the wave vector of the spin-wave which is quantized due to the pinning of the surface spins by surface anisotropy. For such exchange resonance modes, the resonance frequency is always larger than that of the uniform mode, which implies that the resonance occurs at an applied field less than that of the uniform mode. The resonance frequencies depend on the size of the particle through the quantization of the wave vector \( k_p \). In contrast, when the long-range dipolar interactions are stronger than the exchange interactions, as is the case in larger particles, the resulting magnetostatic or Walker modes may be higher or lower in frequency than the uniform mode (indeed, the uniform mode itself can be considered to be a magnetostatic mode), and their frequency is expected to be independent of the size of the particle. For particles of intermediate size, where the exchange and dipolar energies are comparable, one may have spin wave modes whose dynamics are determined by both short range exchange interactions and long range dipolar fields. In this case, numerical simulations show that the resonance frequencies can be both above and below the frequency of the uniform mode, and the resonance frequencies depend in a complicated manner on the size of the particle. To our knowledge, these coupled exchange modes have not been clearly observed in a FMR measurement.
In this Letter, we present FMR measurements on square periodic arrays of submicron size permalloy particles. In addition to the resonance peak expected for a uniform ferromagnetic film, we observe a number of sideband peaks which we associate with spin wave modes in the particles. The position of the peaks can be very sensitive to the relative orientation of the applied external field with respect to the sample. In addition, when the interparticle spacing is very close, the position of the resonance fields show a four-fold symmetry as a function of the relative orientation of the external field, reflecting the underlying symmetry of the periodic lattice. Numerical micromagnetic simulations of the FMR response show that these sideband peaks are associated with coupled exchange and dipole spin wave modes.

The ferromagnetic particle arrays in this study were fabricated by conventional electron beam lithography on thermally oxidized Si substrates. After patterning, the permalloy (Ni_{79}Fe_{21}) films were electron-beam evaporated, resulting in polycrystalline films of thickness $t$ ranging from 70 nm to 90 nm. The arrays have circular ferromagnetic elements, with diameters $d$ ranging from 100 nm to 500 nm. The square lattice constant $a$ was varied from 150 nm to 1 $\mu$m, and the total area of each array was 400 $\mu$m x 400 $\mu$m. Over 40 different arrays were fabricated and measured; only a few representative samples are discussed here. Figure 1 shows a scanning electron micrograph (SEM) of one of the particle arrays. The samples were measured at room temperature in a Varian electron spin resonance cavity spectrometer with an operating frequency of 9.37 GHz, and the orientation of the static dc field could be varied with respect to the sample substrate.

Figure 2(a) shows derivative FMR spectra for a square array of circular permalloy particles with $d = 0.5$ $\mu$m, $t = 85$ nm, and $a = 1.5$ $\mu$m, as a function of the angle of the magnetic field with respect to one axis of the square array (the magnetic field is always in the plane of the substrate). A large resonance peak is observed at $\sim$1400 Oe, which corresponds to the uniform resonance mode. For reference, we also show the equivalent spectrum for a plane permalloy film of thickness 60 nm, with $H_0$ applied in the plane of the film. The peak for the particle
array occurs at a slightly higher magnetic field in comparison to that of the plane ferromagnetic film due to demagnetization effects. As in the plane film, the position of the peak does not shift as a function of the angle of the magnetic field in the substrate plane, which might be expected from the circular symmetry of the array elements. In addition to the peak for the uniform mode, however, the circle array also shows a small peak at lower field (marked by an arrow). As we noted above, similar peaks have been observed in uniform ferromagnetic films and small ferromagnetic particles, where they are associated with standing exchange spin wave resonances in the sample [7,11]. In our case, however, they are associated with exchange spin-wave modes with a small contribution from dipolar interactions, as can be seen by noting their evolution as the distance between the circular particles is reduced. Figures 2(b)-(e) show equivalent FMR spectra for circle array samples evaporated at the same time as the sample of Fig. 2(a). The nominal diameters for the circles (0.5 μm) are the same; the only parameter that is changing is a, which varies from a = 1.5 μm to 600 nm. As the interparticle spacing is reduced, the position of the peak denoted by the arrows move down in field slightly. Since the only interaction that could be changing as the spacing is reduced is the dipolar interactions between particles, this indicates that the resonance frequency of these modes also depends (albeit weakly) on dipolar interactions in the system.

![Image](64x190 to 268x394)

**FIG. 3.** (a) Calculated absorption spectrum for a d =0.5 μm particle with $M_s = 8.47 \times 10^3$ A/m, exchange stiffness $A = 1.3 \times 10^{-13}$ J/m and damping constant $\alpha = 0.05$ (the derivative of this curve is also shown to facilitate comparison to the experimental curves in Fig. 2). In addition to the large peak corresponding to the uniform mode, satellite peaks are also observed on both sides of the uniform mode peak, similar to what is observed experimentally. The precise positions of these peaks do not match the experimental data, since they are very sensitive to the exact values of the parameters used in the calculations. However, the qualitative trend is very similar. In order to demonstrate that the resonance peaks correspond to coupled exchange-dipolar spin-wave modes, we show in Fig. 3(b) the results of another calculation in which all parameters are identical to the calculation of Fig. 3(a), except that the exchange stiffness $A$ is set to zero, effectively eliminating any exchange contribution to the spin-waves. Although the position of the peak corresponding to the uniform mode is not affected, the positions of the peaks corresponding to the spin-wave modes change. In fact, the mode corresponding to the peak below the uniform mode peak essentially disappears, showing that this peak is associated primarily with an exchange spin-wave mode. The resonance fields of the modes above the uniform mode are shifted slightly. With $A = 0$, these spin-wave modes correspond to pure magnetostatic or Walker modes; the difference in the position of these resonances compared to those of Fig. 3(a) show that the resonances of Fig. 3(a) involve

and below the uniform mode resonance, as well as at higher magnetic fields (e.g., at ∼ 2100 Oe). Second, the amplitude of these additional peaks varies as a function of the angle of the magnetic field in the plane of the film. For example, looking at Fig. 2(e) again, the resonance at ∼ 2100 Oe is most clearly developed when the external magnetic field is oriented along a diagonal of the square lattice. Figure 2(f) shows a similar but much more pronounced effect in another circle array sample fabricated in a different run. Third, the field at which the uniform resonance mode occurs oscillates as a function of the angle of the applied magnetic field, as seen most clearly in Fig. 2(e) and (f). This indicates that the dipolar interactions between particles also influence the energy of the uniform mode.

In order to understand the nature of the resonance modes, we have performed time-dependent micromagnetic calculations of the ac response of isolated ferromagnetic particles by numerically solving Eq. (1), using the public OOMMF micromagnetic code solver [12]. The FMR absorption spectrum is obtained by applying a small (∼ 10 Oe) transverse ac field at a frequency of 9.37 GHz at each value of the dc magnetic field, and calculating the amplitude of the steady-state magnetization response at this frequency. The details of the calculation will be discussed elsewhere [13]. Figure 3(a) shows that the resonances of Fig. 3(a) involve

...
a coupling of the exchange and dipolar contributions to
the spin-wave modes.

![Diagram](image)

FIG. 4. Each panel represents the time evolution of the
magnetization distribution through one cycle of the ac field
(at phases $\sim 0, \pi/2, \pi, 3\pi/2$) for a $d = 0.5 \, \mu m$ particle at
magnetic fields corresponding to the peaks labeled 1-3 in Fig.
3(a). (a) peak 1; (b) peak 2; (c) peak 3. $H_0$ is aligned along
the $x$-axis, and the grayscale plot denotes the angle between
the $y$ component of $M$ and the $x$-axis.

Further evidence of the nature of the modes corre-
sponding to the resonance peaks observed in Fig. 3(a)
can be obtained by visualizing the time-dependent mag-
netization at the corresponding values of magnetic field.
The four panels in Fig. 4(a) show the magnetization
distribution during one period of the ac field at the dc
magnetic field corresponding to the uniform mode, de-
noted by the arrow labelled ‘1’ in Fig. 3(a). In order to
make the picture clearer, each arrow in a panel denotes
the magnetization over an area much larger than the cell
size used in the calculation ($5 \, nm \times 5 \, nm$). The uni-
form nature of the mode can be clearly seen in the fact
that the motion of each arrow is almost identical to that
of its neighbors, except for the areas around the edge of
the circle. Figure 4(b) shows a similar evolution for the
resonance peak at $\sim 1000 \, Oe$ (labeled ‘2’ in Fig. 3(a)),
below the uniform mode peak. In this case, the ac re-
sponse of the system consists of small oscillations about
a very non-uniform static magnetization distribution, or
in other words, a spin-wave mode. The fact that this
mode almost completely disappears when the exchange
stiffness $A = 0$ suggests that the major contribution to
the energy of this mode comes from the standing ex-
change spin-wave, as discussed by Kittel. Figure 4(c)
shows the time evolution over one period of the ac field
for the resonance peak at $2500 \, Oe$, labeled ‘3’ in Fig.
3(a). As can be seen, the nature of this mode is quite
different from either the uniform mode or the primar-
ily spin-exchange mode we have just discussed. In this case,
the magnetization in the center of the particle is mostly
static and aligned along the direction of the magnetic
field; the response of the particle is confined primarily
to small oscillations of the magnetization near the edges.
Due to shape demagnetization effects, the magnetization
near the edges is non-uniform, and will give rise to an
exchange contribution to the mode energy. We believe
it is also these edge effects which give rise to the small
dipolar contribution to the spin-wave mode labeled ‘1.’
The confinement of the non-uniform magnetization to the
edges of the circle also suggests why these modes may be
influenced strongly by the dipolar field arising from an-
other ferromagnetic particle placed in close proximity, as
we have seen in our experimental results. In principle,
one could model an array of circular particles, but this
involves the use of much greater computer resources than
we have at our disposal at present.

In summary, we have measured the FMR response
of arrays of circular ferromagnetic particles. In addi-
tion to the uniform mode of precession, the FMR spec-
tra shows resonances corresponding to non-uniform spin-
wave modes of precession at values of magnetic field both
above and below the peak corresponding to the uniform
mode, some of which are strongly influenced by dipolar
interactions between the particles. Our experimental re-
sults and numerical simulations show that these modes
correspond to coupled exchange and dipolar spin-waves.

We thank Anupam Garg for useful discussions, and
Robert Tilden for use of computer facilities. This
work was supported by the Army Research Office
through DAAD19-99-1-0339, and by the David and Lu-
cile Packard Foundation.

[1] Y. Martin and H.K. Wickramasinghe, Appl. Phys. Lett.
50, 20 (1987).
[2] See, for example, Ultrathin Magnetic Structures II, edited by B. Heinrich and J.A.C. Bland (Springer, Berlin, 1994).
[3] K. Wago, O. Züger, R. Kendrick, C.S. Yannoni, and D. Rugar, J. Vac. Sci. Technol. 14(2), 1197 (1996).
[4] See, for example, A. Aharoni, Introduction to the Theory of Ferromagnetism (Oxford University Press, New York,
1996).
[5] C. Kittel, Introduction to Solid State Physics, 7th Edition
(John Wiley and Sons, New York, 1996).
[6] C. Kittel, Phys. Rev. 110, 1295 (1958).
[7] M.H. Seavey, Jr, and P.E. Tannenwald, Phys. Rev. Lett.
1, 168 (1958).
[8] L.R. Walker, Phys. Rev. 105, 390 (1957).
[9] P.A. Voltairas, D.I. Fotiadis, and C.V. Massalas, J. Appl.
Phys. 88, 374 (2000).
[10] R. Arias and D.I. Mills, Phys. Rev. B 63, 134439 (2001).
[11] Ph. Toneguzzo, O. Acher, G. Viau, F. Fiévet-Vincent,
and F. Fiévet, J. Appl. Phys. 81, 5546 (1997).
[12] M.J. Donahue and D.G. Porter, URL:
http://math.nist.gov/oommf
[13] S. Jung, J.B.Ketterson and V. Chandrasekhar, unpublished.