I. INTRODUCTION

The physical properties of ferromagnetic particles continue to be an active area of fundamental experimental and theoretical research. Advances in lithographic and measurement techniques are now permitting some of the basic tenets in the field of small particle classical magnetism to be critically tested. For example, measurements of a single acicular γ-Fe₂O₃ particle and electron beam fabricated rectangular permalloy particles are not consistent with the Néel–Brown theory of thermally assisted magnetization reversal over a simple potential barrier. Usually this theory is the starting point in the analysis of more complex particulate media which have a distribution of sizes, shapes, and interactions. Magnetization reversal in elongated particles has also recently been re-examined theoretically. In idealized filaments Braun finds that spatially localized magnetization fluctuations increase the switching rates and hence reduce the measured coercivities at finite temperature relative to the Néel–Brown theory. Real samples, of course, contain defects, ends, and surfaces that are not considered in this work but which are expected to play an important, if not dominant, role in the physics of magnetization reversal. Of fundamental interest is the mechanism of reversal in real particles—whether it is a classical coherent (Néel–Brown, Braun) or incoherent mode or, alternatively, heterogeneous nucleation and growth. We have set out to study smaller noninteracting particles in which the competing effects can be highlighted by systematically varying the particle dimensions.

It is the purpose of this article to show how this is accomplished in scanning tunneling microscope (STM) nanolithographically produced small particle systems. STM and chemical vapor deposition techniques have been used to fabricate nanometer scale diameter (~25 nm) iron particles with a range of shapes from nearly isotropic to filamentary. Deposits are fabricated directly in the active area of a newly developed high sensitivity Hall magnetometer. With this device hysteresis loops of dilute particle arrays comprising 100–600 particles (~×10⁻¹¹ emu) have been measured at low temperature. Moreover, the magnetic characteristics have been studied for a variety of particle sizes and shapes. In particles of intermediate aspect ratio (2.2:1 height:diameter) and diameter the largest coercive force is observed. This observation is not consistent with the well-known classical coherent or incoherent modes of reversal. Further, from the hysteresis loops and array geometry we estimate the particle magnetization and interparticle interaction strength.

We begin with a brief review of the materials fabrication technique and characterization. This is followed by a discussion of the magnetic measurements and analysis on particles of systematically varied geometry.

II. FABRICATION

Iron particles are formed by using a STM to decompose iron pentacarbonyl [Fe(CO)₅] which is metered into the mi-
croscope’s ultrahigh vacuum chamber ($P=2 \times 10^{-10}$ T). To initiate the growth the substrate-tip bias is raised to 15 V and a current of 50 pA maintained in the presence of 30 μTorr of Fe(CO)$_3$. The STM feedback loop is active and maintains a constant current and thus constant height between the tip and growing deposit. When the deposit has reached the desired height above the surface the tip is retracted to stop the growth. The tip is moved to another location and the process repeated to form arrays.

Characterization by both Auger electron spectroscopy (AES) and transmission electron microscopy (TEM) indicate that relatively pure iron deposits are formed under these conditions. TEM shows that these consist of a polycrystalline bcc iron interior surrounded by a contamination coating in which the grain size is approximately the inner core diameter. AES reveals greater than 70 at. % Fe with a carbon remainder. The fact that the bcc phase is formed is evidence for greater purity of the deposits in their interior since the equilibrium phase of the Fe-C system above 0.4% carbon is fcc.$^8$

The scanning electron micrographs in Figs. 1(A) and 1(C) show the extremes in dimensions we have fabricated and studied. The particle geometry, with the long axis perpendicular to the substrate surface, is highlighted in the oblique view presented in the micrograph. Note that from Figs. 1(A) and 1(C) the particle diameters progressively decrease from 42 to 17 nm while the ratios of height to diameter increase from 1 to 6. Although these measurements serve as a basis for comparing particle dimensions, contamination built up during observation as well as the finite resolution of the SEM cause systematic overestimates of the actual particle size.$^9$ The interparticle distance is approximately 130 nm and was chosen to minimize, as much as possible, the effect of interparticle interactions while building up enough overall moment for measurement. Magnetic measurements have been made on these samples as well as on some of intermediate dimensions.

**III. MEASUREMENTS**

Measurements were made using a novel high sensitivity magnetometer based on the Hall response in a semiconductor heterostructure. In contrast to integrated superconducting quantum interference device microsusceptometers,$^{10}$ this device allows for systematic investigations over a wide range of applied fields and temperatures. A high mobility GaAs/Ga$_{0.7}$Al$_{0.3}$As two-dimensional hole gas sample ($n_{2D}=3 \times 10^{11}$ cm$^{-2}$, $\mu(5 \text{ K})=10^7$ cm$^2$/V s) was wet chemically etched into the form depicted in Fig. 2. With the bridge properly balanced, the resulting output voltage $V$ is proportional to the sample contribution to the magnetic induction. This contribution can then be calculated using the measured Hall coefficient so that $\Delta B=V/R I$ where $R$ is the Hall coefficient ($\sim 0.2 \text{ O} / \text{G}$) and $I$ is the measurement current. Typically we use a 6.5 Hz ac current ($\sim 1 \mu A$/rms) and lock-in detect the difference signal $V$.

In practice, a difference voltage is present even in the absence of a magnetic sample due to small variations in the Hall crosses (typically $\sim 0.1\%$). This imbalance results in a signal proportional to the applied magnetic field which is minimized by adjusting the ratio $I$ to $I'$ at high field ($H \gg H_c$, the coercive field).

The large Hall response ($\sim 0.2 \mu V/G$) in combination with good coupling of small samples to the device results in an excellent spin sensitivity. For example, the observed field noise of 0.1 G/Hz$^{1/2}$ (at 0.1 Hz, 5 K) in a 2 $\mu m^2$ device implies a spin sensitivity of $10^{-14}$ emu/Hz$^{1/2}$. Typically, signals from the STM arrays are 10 times this noise level. In addition, the sensor works over a large range of magnetic field and temperature. At low temperature, ballistic transport (on the scale of the magnetic arrays) reduces the responsiveness. The quantum Hall effect and the associated nonlinear dependence of Hall voltage on magnetic field also changes the response at low temperature and high fields. Both the decreasing hole mobility and thermally activated switching of defects in GaAs cause the noise to increase with temperature. We have successfully used this magnetometer from 1 to 80 K with some decrease in performance with increasing temperature ($T>50$ K).

An array of particles of intermediate size (those in Fig. 1(B), referred to as sample (B)) placed in the active area of...
the magnetometer is shown in Fig. 3. An external field is applied perpendicular to the plane of the device and hence parallel to the long axis of the particles. Hysteresis loops are measured starting from a saturating field, ramping at a constant rate to the opposite field polarity and then back. Figure 4 shows the difference in induction, which is proportional to the sample magnetization, plotted versus the external field. Measurements were made at different temperatures and ramp rates. Under these conditions the coercive field has only a slight dependence on the measurement time and temperature.

and is approximately 2.7 kOe. The magnetization reversal occurs over a range ±0.4 kOe about the value. In Fig. 4(d), as a check, the same measurement was made using two reference crosses, neither of which contained a sample. This shows no hysteresis and only a small deviation from a constant value.

Magnetic measurements were made on arrays (A), (B), and (C) in Fig. 1 in order to study the dimensional dependence of the magnetic properties (Fig. 5). The shapes of the hysteresis loops change significantly in going from the nearly isotropic particles of sample (A) to the filamentary particles in (C). Most notably, the loops are increasingly square with more abrupt magnetization transitions. The change in sample magnetization also decreases due both to the geometry of the samples as well as to the decreasing moment per particle. The coercivity initially increases in more anisotropic particles [(A) to (B)]. Surprisingly, this is followed by a decrease in the more filamentary particles of sample (C). Samples intermediate in dimensions to (B) and (C) appear to confirm this trend.

The loop shape for sample (A) is close to that expected from the Stoner–Wohlfarth theory for noninteracting uniaxial particles with randomly oriented easy axes. For instance, the remanence is 0.46 times the saturation magnetization close to the 0.5 predicted by SW. The coercivity is 640 Oe. Within this model the anisotropy field is \( H_{\text{a}} \sim 0.48 \) or 1.33 kOe. Small deviations from isotropic shape would account for this entire anisotropy although a crystalline contribution might also be present (bcc Fe\(^{6-540}\) Oe). The demagnetization factors of a prolate ellipsoid of iron with axial ratio 1.17 is sufficient to explain this and within the range of measured particle asymmetries \( c/a = 1.3 \pm 0.3 \), Fig. 1(A). For the oriented elongated particles [samples (B) and (C)] increasingly
previously mentioned, it seems appropriate to assume that
pared to the 29 nm diam measured from Fig. 1(B). This
The results for the other samples are summarized in Table I.

where $m$, the magnetic moment per particle, is the quantity
measurements. Taking the measured magnetization of bee Fe
approximately 10 smaller than an estimate based on the bulk
Hall cross can be performed analytically and then summed
the array geometry and position in the Hall bar by a spatially
and $z$ the distance from the dipole's center to the heterointerface. The integral over the rectangular surface $S$ of the
area is given by

$$
\Delta B = \frac{2m}{S} \sum \int_{S} \frac{1}{\left[ z^2 + (r - r_i)^2 \right]^{3/2}} \times \left( 1 - \frac{3z^2}{z^2 + (r - r_i)^2} \right),
$$

where $m$, the magnetic moment per particle, is the quantity
interest; $r$ is a vector in the plane of the surface; $r_i$ the
position of the $i$th particle; $S$ the surface of the Hall cross,
and $z$ the distance from the dipole's center to the heterointerface. The integral over the rectangular surface $S$ of the
Hall cross can be performed analytically and then summed
particles numerically to solve for $m$. This calculation
approximates the actual nonuniform field distribution from
the array geometry and position in the Hall bar by a spatially
homogeneous average field in estimating the moment. For sample (B), we find $m=5.4 \times 10^{-15}$ emu. This is a factor of
approximately 10 smaller than an estimate based on the bulk
moment of Fe and our observations of the particle size. As
previously mentioned, it seems appropriate to assume that
the magnetic volume is smaller than that found from SEM
measurements. Taking the measured magnetization of bcc Fe
(1700 emu/cm$^3$) gives a magnetic volume of $3 \times 10^{-17}$ cm$^3$ or
equivalently a magnetic core only 9 nm in diameter, compared
to the 29 nm diam measured from Fig. 1(B). This
interior size is consistent with TEM observations on similar
deposits. Within this analysis, the aspect ratio of the magnetic
volume would also be greater than the measured value.
The results for the other samples are summarized in Table I.

From the particle moment the strength of the dipolar
interactions between particles can be estimated. For a square
lattice of oriented dipoles with spacing $a$, the interparticle
interaction field due to nearest neighbors is $4m/a^3$. For
sample (A) this is 20 G. Substantial contributions result from
the long range nature of the dipole interactions. Including
oriented neighbors out to four lattice spacings (48 particles)
results in a field of 40 G. This is still much less than the
anisotropy field and will play a role only in the tails of the
magnetization transition. Interactions are less important in
samples (B) and (C) due to their smaller moment per par-
ticle.

IV. DISCUSSION

Changes in the coercive force with dimensionality, notably
the decrease in high aspect ratio smaller diameter par-
ticles, are not consistent with conventional coherent\textsuperscript{10} or in-
coherent modes of spin reversal.\textsuperscript{5} For instance, the curling
mode, which is applicable only in larger diameter particles,
predicts an increase in coercive field with decreasing diam-
eter. In both coherent reversal and fanning models the coerc-
ive field increases with aspect ratio. For reference, coherent
reversal in an iron particle of axial ratio 2.2 [like sample
(B)], would require a field of 5.7 kOe—twice the observed
value. Fanning, which postulates decoupled grains, at least
can account for the magnitude of the observed switching
field.

Recently, Braun has considered the role of nonuniform
magnetization fluctuations in reducing the coercivity at finite
temperature in elongated particles.\textsuperscript{6} It is therefore worth es-
imating whether this theory applies to our measurements.
Fluctuation effects are important when the thermal energy is
comparable to the barrier height. For uniform coherent rever-
sal, at zero field, the barrier is $KV$, the anisotropy volume
product. To be specific, for sample (C) this is $10^4$ K. At low
temperature this implies that such reversal cannot occur until
the applied field is very close to the intrinsic (zero tempera-
ture) coercivity. For example, at 5 K the particles switch
when $H/H_c \sim 0.9$, with our measurement times. In Braun's
theory, the energy is no longer proportional to the volume
but the cross-sectional area of the particle, $8JK/c \ A$. This
is the energy density of a domain wall times the cross-
sectional area, $A$. $J$ is the exchange constant and $c$ the lattice
spacing. An estimation of this energy for sample (C) is also
$10^4$ K. Thus spatially nonuniform reversal requires the same
amount of thermal energy as the uniform case due both to the
large exchange and anisotropy energies. Only in far more
filamentary particles does the nonuniform mode dominate.
These estimations suggest that fluctuations cannot account
for the observed reduction in coercivity.

As an alternative to the above models, the particle ends,
surfaces, and defects might play an important role in deter-
mining the magnetic properties. For example, large demag-
netization fields such as occur at the particle ends would
favor a heterogeneous nucleation and growth of reversed do-
 mains as occurs in bulk ferromagnets.\textsuperscript{7} Reversal would con-

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**TABLE I. Summary of particle properties.**

<table>
<thead>
<tr>
<th>Sample</th>
<th>No. of particles</th>
<th>diam (nm)</th>
<th>c/a</th>
<th>$m$ (emu/particle)</th>
<th>$H_c$ (Oe)</th>
<th>diam (nm)\textsuperscript{b}</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>500</td>
<td>42 ± 7</td>
<td>1.3 ± 0.3</td>
<td>$1.0 \times 10^{-14}$</td>
<td>1324\textsuperscript{a}</td>
<td>21</td>
</tr>
<tr>
<td>B</td>
<td>600</td>
<td>29 ± 2</td>
<td>2.2 ± 0.3</td>
<td>$5.4 \times 10^{-15}$</td>
<td>2700</td>
<td>9</td>
</tr>
<tr>
<td>C</td>
<td>100</td>
<td>17 ± 1</td>
<td>5.8 ± 0.5</td>
<td>$1.6 \times 10^{-15}$</td>
<td>2050</td>
<td>4</td>
</tr>
</tbody>
</table>

\textsuperscript{a} Anisotropy field determined from the measured coercivity and the SW model.
\textsuperscript{b} Magnetic core diameter inferred from the magnetic moment and particle shape.
sist of either the localized nucleation of a reversed domain and its subsequent propagation or the movement of an existent closure domain through the particle. Moreover, defects, such as grain boundaries, lower the barrier to domain nucleation. As the energy of a domain wall is proportional to its area these scenarios are qualitatively consistent with the observed decrease in coercive force in smaller diameter particles.

In summary, we have demonstrated the ability to fabricate and measure nanometer scale ferromagnets. Magnetic structures with well-defined geometries have been coupled to an electronic system. This is the basis for a novel high sensitivity magnetometer which relies on the Hall effect in a semiconductor heterostructure. In addition to investigations of magnetic nanostructures, this combination enables studies of the effect of local magnetic interactions on electronic properties and transport in semiconductors.

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8See for example, Binary Alloy Phase Diagrams, 2nd ed., edited by T. B. Massalski (Scott, Materials Park, 1990), pp. 842–848.
9See the TEM photo in Fig. 3 of Ref. 7.