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Citation: Journal of Applied Physics **81**, 4331 (1997); doi: 10.1063/1.364760
View online: http://dx.doi.org/10.1063/1.364760
View Table of Contents: http://scitation.aip.org/content/aip/journal/jap/81/8?ver=pdfcov
Published by the AIP Publishing

Assembly and imaging of submicron ferromagnets in GaAs (invited)

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Submicron room-temperature ferromagnets are formed in GaAs semiconductors through a simple process of ion implantation and subsequent heat treatment. The ferromagnetic structures are studied using magnetic force microscopy. Magnetization switching of single-domain particles has been directly imaged in applied magnetic fields. In order to understand the images of magnetic structures taken under external fields, we have characterized the magnetic force microscopy (MFM) probes by imaging microfabricated current-carrying strips in applied magnetic fields. Patterned micrometer scale lines containing submicron magnetic structures on GaAs are fabricated using lithography in conjunction with broad beam ion implantation.
[S0021-8979(97)36208-2]

I. INTRODUCTION

Incorporation of ferromagnetic structures into semiconductors is of great scientific and technological interest. Such materials are needed not only for exploring novel spin-dependent phenomena in semiconductor based structures, but also for developing new magneto-electronic devices utilizing these phenomena.[1] Molecular beam epitaxy (MBE) is often required for tailoring the ferromagnet/semiconductor structures in these applications.[2] We reported earlier, however, that submicron ferromagnets can be successfully fabricated in GaAs by means of a simple process of ion implantation and subsequently heat treatment.[3,4] Here we report further study of magnetic properties using magnetic force imaging and determination of magnetic force microscopy (MFM) tip properties. We also describe recent attempts to ultimately fabricate one-dimensional ordered ferromagnetic structures in GaAs.

II. SAMPLE PREPARATION AND CHARACTERIZATION

Most of the semiconductor samples for this magnetization study are grown by MBE on semi-insulating GaAs substrates, and consist of 500 nm undoped GaAs epitaxial layers grown atop 50 nm AlAs and a 500 nm GaAs buffer. These structures are then exposed to uniform Mn^+ ions at energies of 50 and 200 keV and doses ranging from 1×10^{14} to 5×10^{16} Mn^+/cm^2. Subsequent rapid thermal annealing (at temperatures from 800 to 920 °C for durations ranging from 5 s to 20 min) is needed for the formation of submicron ferromagnets on a GaAs surface. The MBE grown structures are purposely designed to facilitate magnetization measurements, as removal of the substrate by selectively etching AlAs slab reduces the diamagnetic background. However, the epitaxial growth is not essential to the formation of submicron ferromagnets in GaAs and the same results can be achieved with semi-insulating GaAs materials as well.

Transmission electron microscopy and atomic force microscopy[3,4] revealed that the precipitates formed at the surface range from 200 to 400 nm in diameter, with densities from 3×10^7 to 1.5×10^8 cm^{-2}. The precipitates are composed of Ga and Mn at a ratio about 60:40. Detailed analysis of electron diffraction patterns indicates that a large percentage of precipitates exhibits quasicrystal and quasicrystallike symmetry. Superconducting quantum interference device (SQUID) magnetometry results show that these GaMn precipitates are ferromagnets with a Curie temperature well above 400 K. In a known Ga–Mn binary phase diagram, GaMn phases at or near this composition are ferromagnetic phases with lower Curie temperatures or simply nonmagnetic phases.[5] The ferromagnetism of the precipitates may be correlated with the quasicrystal nature of the precipitates, which are the products of highly nonequilibrium processes. Note that in this method much higher Mn concentration (about three orders of magnitude above the equilibrium solubility) is achieved.

III. MAGNETIC FORCE MICROSCOPY AT ZERO MAGNETIC FIELD

MFM is a powerful tool for studying submicron room temperature ferromagnetic structures. Since GaMn particles are formed on the GaAs surface, MFM is particularly useful for probing these local magnetic moments. A typical MFM image of an unmagnetized sample is shown in Fig. 1, taken with a vertically magnetized MFM tip (magnetized perpendicular to the sample surface). Many particles show very strong magnetic contrast, which results from magnetic interactions between the MFM probe and the opposite poles on ferromagnetic particles. Particles such as *d* and *e* appear to have less complicated magnetic contrast patterns than particles *a*, *b*, and *c*. Note that the other group of particles shows weaker magnetic contrast (~30 times smaller in magnitude). This can be caused by lower coercivity and smaller magnetization of the magnetic particles, due to variations in GaMn relative concentration.

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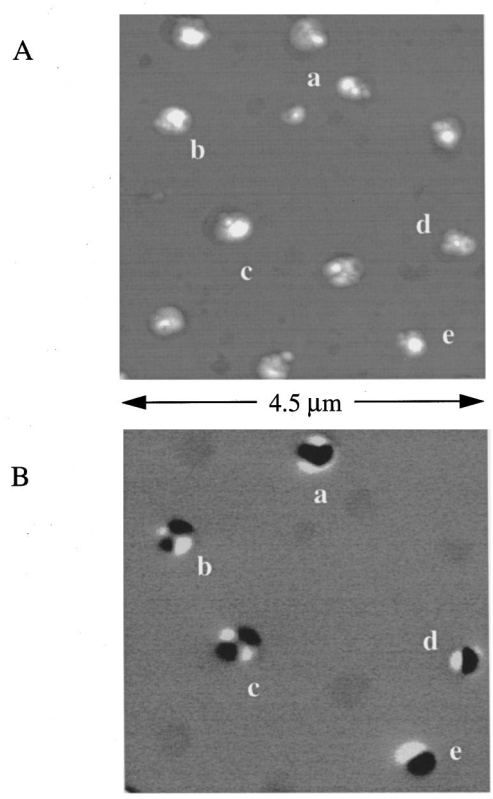

FIG. 1. Topographic (A) and magnetic force (B) images taken on an unmagnetized GaAs sample (implanted with 1×10^{15} Mn^+/cm^2, annealed at 920 °C for 60 s). The particles are about 400 nm in diameter and about 200 nm above the GaAs surface. MFM tip is magnetized perpendicular to the sample surface and the tip-sample separation is about 50 nm.

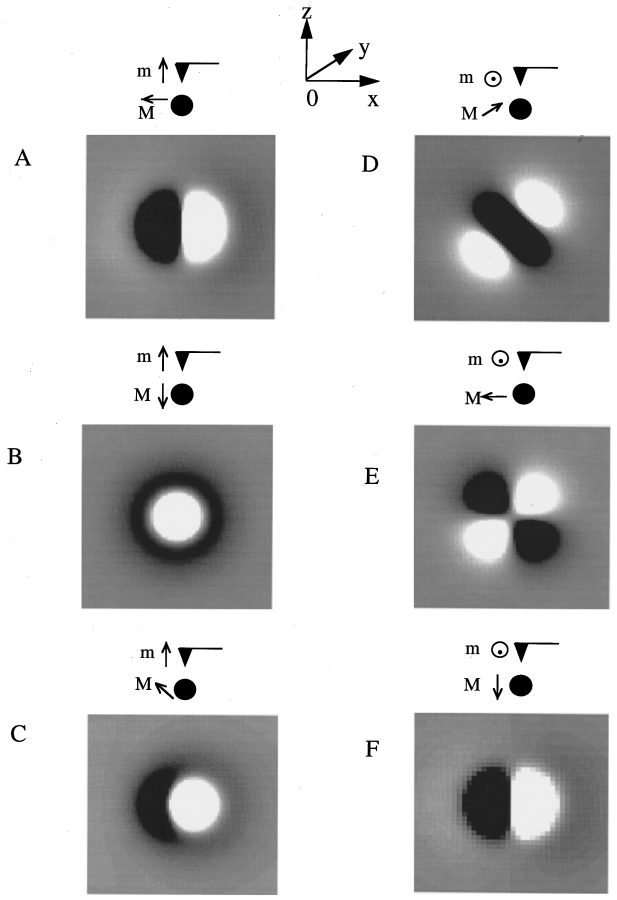

FIG. 2. Calculated magnetic force images for a single-domain particle with different moment orientations, **M**, with respect to a tip moment **m**. On the left, **m** is perpendicular to sample surface; on the right, **m** parallel to sample surface. (A) **M** parallel to surface, (B) **M** perpendicular to surface, (C) **M** canted 45° from surface normal, (D) **M** parallel to surface but at 45° with **m**, (E) **M** parallel to surface but perpendicular to **m**, and (F) **M** perpendicular to surface.

A coated MFM tip can often be regarded as a magnetic point dipole,[6] and the MFM response measures the second derivative of the magnetic field component projected in the direction of the tip moment vector. For a single-domain particle, whose magnetization is uniform, the derivative of the stray field can be calculated and is plotted in Fig. 2. Depending on the relative angle between the tip moment, **m**, and the single-domain particle moment, **M**, the calculated field derivative pattern varies from (A) to (F). The vertical field profiles (left column) are generally less complicated than horizontal field profiles (right column). From the above analysis, we can conclude that the multi-component MFM patterns, such as particles *a*, *b*, and *c* observed in Fig. 1, can only be produced by multi-domain particles. Although images taken with a horizontally magnetized tip sometimes cannot unambiguously distinguish single-domain from multi-domain particles, for example, (D) and (E) in Fig. 2, examining the vertical field profile with a vertically magnetized MFM tip can provide a definitive answer as to the magnetic state of particles. This fact also underscores the importance of knowing the magnetic state of MFM tips in order to interpret images correctly.

As reported earlier,[4,7] an unmagnetized sample usually contains both single- and multi-domain GaMn particles (such as particles *d* and *e*). Upon application of a large magnetic field, domain walls within the multi-domain particles are swept out, and magnetic moments of single-domain particles are rotated to minimum energy directions. According to our earlier MFM studies, after initial magnetization and removal of the external field, the magnetic moments are found to be aligned preferentially along the [100] crystallographic directions of the GaAs substrate.

To check the stability of the submicron ferromagnets, magnetization of an implanted (5×10^{16} Mn^+/cm^2) and annealed (920 °C for 60 s) sample is measured again after it has been exposed to air for over 12 months. The magnetization results are reproduced, indicating that magnetic property of the submicron GaMn precipitates are stable in an ambient environment.

IV. MFM PROBE CHARACTERIZATION USING CURRENT STRIPS

MFM is usually employed to image unmagnetized or remanent states of magnetic samples at zero magnetic field. In these cases, MFM tips are often magnetized with a vertical field; and are assumed to remain magnetized in the vertical direction. To investigate intermediate states between different remanent states, and reveal a dynamic single-domain switching process, sometimes it is desirable to do

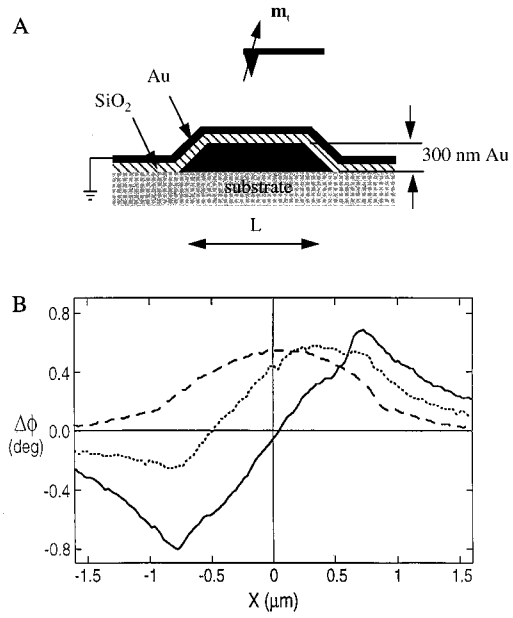

FIG. 3. (A) Cross section of a current carrying strip for studying magnetic properties of MFM probes. Micron and submicron ($L=1$, .5, and .2 μm) wide Au lines are made using e-beam lithography, and separated from a top Au layer by a SiO_2 dielectric layer. (b) MFM response (phase shift) showing profiles which are antisymmetric for a tip magnetized vertically (solid curve), symmetric when magnetized laterally (dashed curve), and mixed for a canted moment (dotted curve). For the lateral case, a horizontal field (1.5 kOe) was applied continuously.

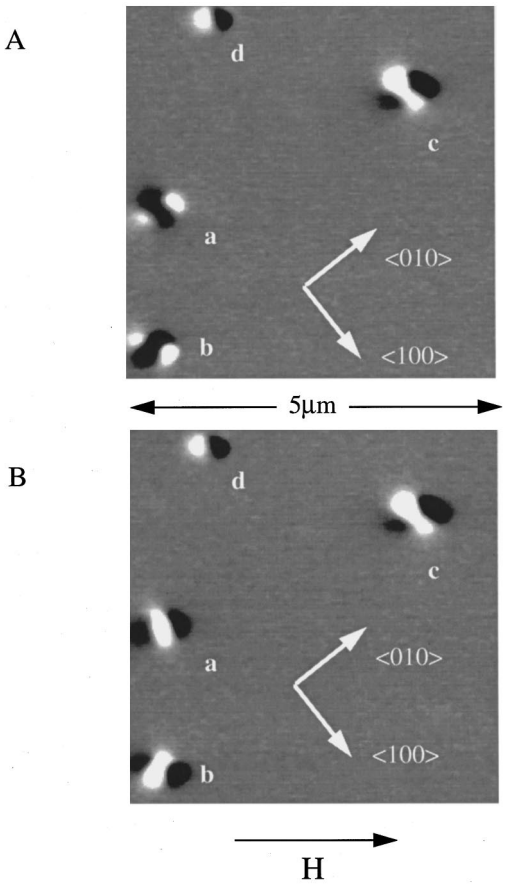

FIG. 4. MFM images of single-domain GaMn particles (implanted with 1×10^{15} Mn^+/cm^2, and annealed at 920 °C for 60 s.) at 2 kOe (A) and 5 kOe (B) lateral fields. The sample was previously magnetized in an in-plane field to the left. Particle *a*, *b*, and *c* correspond to in-plane moments, and *d* corresponds to a moment normal to the sample surface.

real-time imaging in changing magnetic fields. However, both the sample under examination and the MFM tip are subject to the external field. Consequently, the tip moment orientation can be rotated away from its original vertical direction when a horizontal field is applied. As a result, interpretation of MFM images becomes a complicated task, even for single-domain particles, for the reasons described in the last section. One must know how a MFM tip itself responds to the external magnetic field in order to understand the evolution of images in changing fields.

Since a MFM tip carries only a very small active magnetic volume, typically about 10^{-12} emu, it is extremely difficult to directly determine its magnetic properties using conventional magnetometry. We have fabricated miniature current strips using electron beam lithography for characterizing the MFM tips.[6] Since the field and field derivative profiles generated by the current strips can be calculated for given tip moment orientations, information regarding the magnetic state of the MFM tip can be extracted from the magnetic force contrast of the current strips. Moreover, an applied field does not alter the field profile of the current strip, and thus the field dependence can be obtained from performing MFM imaging in a changing field, yielding hysteresis loops of the tip moment. Detailed descriptions of the experiments have been reported elsewhere.[6] Figure 3(A) shows the cross section of a current strip on an insulating substrate. The width of the conducting layer (Au), L, ranges from .2 to 1 μm, and the height is about 300 nm. To shield electrostatic fields from the current-carrying layer which would otherwise exert a force on the MFM tip, a top conducting layer is grounded, and separated from the Au lines with a SiO_2 dielectric layer. When a tip is magnetized in known directions, the MFM does give expected responses with expected symmetries, as shown in Fig. 3(B).

Various experiments have been carried out to obtain magnetic hysteresis loops of different tips in both vertical and horizontal fields. Coercivity and saturation field are the two most important quantities obtained from such measurements.[6] Knowing these parameters, one can select appropriate tips for various applications. MFM can operate in the field range where the tip magnetization stays constant when the applied field varies, so that any change in contrast is solely due to the sample. For GaMn particles, the coercive field is about several kOe, therefore CoCr coated pyramidal tips are satisfactory, because the tip's coercive field (about 400 Oe) is well below the particle's coercive field.

V. MFM STUDY OF GaMn PARTICLES IN APPLIED FIELDS

An electromagnet has been built for studying dynamic switching of magnetization during MFM imaging. The magnet provides an in-plane field up to 8 kOe. When operating in constant field mode, thermal drift is insignificant for fields below 5 kOe. Figures 4(A) and 4(B) show MFM images of GaMn single-domain particles at 2 and 5 kOe, respectively.

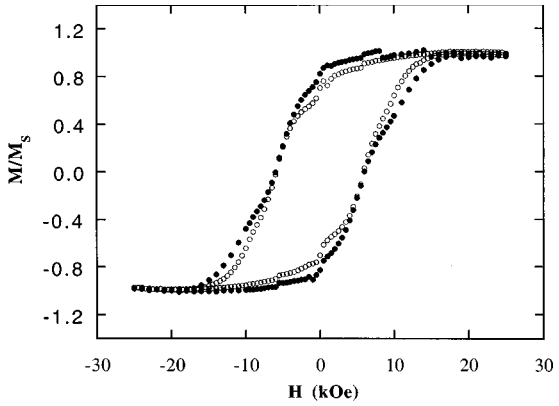

FIG. 5. Magnetic hysteresis loops at 50 K for a magnetic field parallel (open circles) and perpendicular (solid circles) to GaAs sample surface (implanted with 5×10^{16} Mn^{+}/cm^{2} and annealed at 920 °C for 60 s). The diamagnetic background has been removed for both curves.

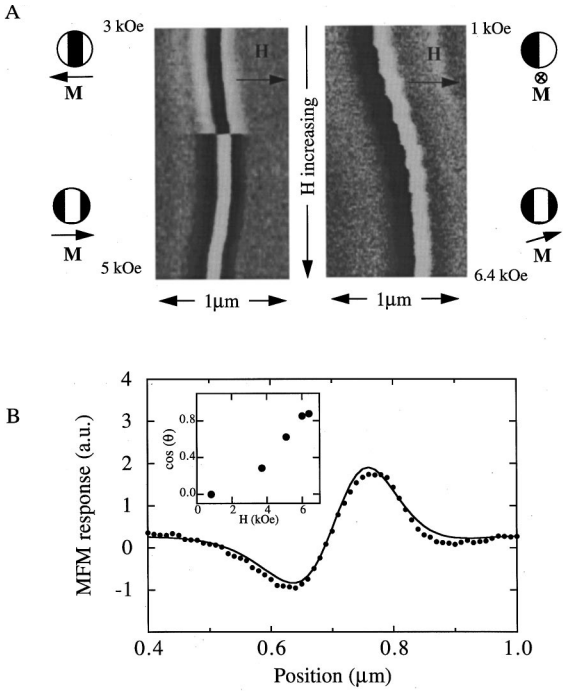

FIG. 6. (A) Magnetic force images taken in changing in-plane magnetic fields for moments parallel (left) and perpendicular (right) to the field. (B) Cross section of MFM data (solid circles) and fit (solid curve) for the moment perpendicular to the field (at 6.4 kOe). Inset shows M/M_s as a function of the applied field.

The sample was previously saturated with a very large field along the ⟨110⟩ direction of the GaAs substrate, and then the in-plane field is reversed to the ⟨110⟩ direction. Note that at and above 2 kOe, the MFM tip is already aligned with the external field direction and the in-plane moments, *a*, *b*, and *c* resemble the pattern shown in Fig. 2D. Under the 2 kOe field, the magnetic force contrast of particle *c* has inverted, and magnetization has already reversed from its original orientation. Meanwhile, other in-plane moments *a* and *b* undergo gradual rotation toward the field direction, and particle *d* remains normal to the surface. At 5 kOe, magnetization of both *a* and *b* reverses, and, in addition, magnetization of *a*, *b*, and *c* is more tilted toward the applied field direction. For single-domain particles, both the sudden reversal and gradual rotation of the magnetization contribute to general magnetic hysteresis behavior.

Imaging in different areas over many particles demonstrates that there is a large variation in switching field among particles with the *same* easy axis orientation, and this variation can be as large as a factor of 3. For a group of nearly spherical particles, a small variation in detailed shape could result in relatively large fluctuations in the switching field $H_c(H_c\sim(N_b-N_a)M$ in the Stoner–Wohlfarth picture,[8] where N_b and N_a are demagnetizing factors in the short and long axes,[9] and M is the magnetization). However, experiments indicate that the magnetic easy axis orientation is strongly correlated with the crystallographic directions of GaAs substrate, rather than random variations in particle shape. The fluctuations may be caused by other factors, such as surface or strain-related anisotropy. At present, the underlying physics is not fully understood.

Note that there is very little change in contrast for particle *d* (with a vertical moment) between 2 and 5 kOe, indicating a very high switching field. One may suspect that some anisotropy might cause the difference in switching behavior between the in-plane and normal magnetic moments. To resolve this issue, we have measured magnetic hystersis loops for the field parallel and perpendicular to the sample surface and the results are shown in Fig. 5. The samples are thinned down to about 0.5 mm thick, and *a* large diamagnetic background for each curve has been removed. The results show that there is little anisotropy between the in-plane and normal magnetic moments.

The dynamical switching process of individual particles can be followed by doing MFM imaging *in situ* in a changing magnetic field. Two extreme cases are demonstrated in Fig. 6(A). On the left, an in-plane moment is chosen, and the initial magnetization points opposite to the applied field direction. On the right, the moment is normal to the surface, therefore perpendicular to the applied field direction. MFM tip scans repeatedly over the same particles while the magnetic field is swept.

For the particle whose easy axis is parallel to the applied field direction, an abrupt flip of magnetization is observed in real time. This transition corresponds to a crossover from one state to the other when the magnetization overcomes a certain energy barrier. When the applied field is perpendicular to the easy axis, the magnetization gradually rotates toward the field direction. As clearly seen in the right image, the contrast evolves from an initially antisymmetric to partially symmetric pattern at about 6.4 kOe. The deviation from the initial orientation is shown in Fig. 6B. The solid curve is a fit to the second derivative of the lateral component of the dipolar field generated by the particle. This fit extracts an angle between the moment and the applied field direction, in this case approximately 28°. The fit is done for each field and cosine of this angle (or M/M_s) is plotted as the applied field in the inset. The linear field dependence of the M/M_s is also in agreement with the Stoner–Wohlfarth description of magnetization coherent rotation of single-domain particles.[8]

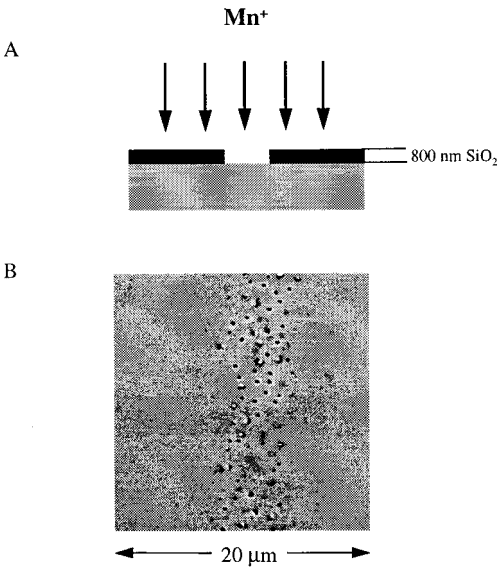

FIG. 7. (A) GaAs sample with SiO_2 mask for producing submicron GaMn particles within micrometer scale narrow lines. GaAs is implanted with 5×10^{16} Mn^+/cm^2, then SiO_2 mask is removed before thermal annealing (920 °C for 60 s). (B) MFM images showing a patterned 5-μm-wide line. MFM tip magnetization is vertical and the sample is unmagnetized.

VI. PATTERNED MAGNETIC STRUCTURES IN GaAS

Submicron ferromagnetic GaMn formation has been demonstrated in GaAs through broad ion beam implantation and subsequent heat treatment. The GaMn particles are distributed everywhere in the implanted region. This method makes it possible to do detailed structural characterization and magnetic property studies. However, the spatial location of particles relies entirely on the nucleation of GaMn precipitates and Mn^+ ion diffusion process. In order to ultimately make spatially ordered magnetic structures in GaAs for more controlled experiments, we have attempted to extend this method to fabricate submicron ferromagnetic GaMn particles along a narrow line using SiO_2 hard mask during Mn^+ ion implantation. As shown in Fig. 7(A), the SiO_2 mask is about 800 nm thick, and features on the mask are about 5 μm wide and 40 μm long. During the implant, Mn^+ ions do not penetrate the SiO_2 layer, and therefore only the exposed areas receive Mn^+ ions. Subsequent particle formation then occurs selectively in these areas.

An example shown in Fig. 7(B) is a magnetic force image taken on such a patterned GaAs. This sample receives 5×10^{16} ions/cm^2, and has been annealed at 920 °C for 60 s. Clearly, GaMn particles are formed only in the exposed area (5 μm wide). Particle size and density are consistent with those found in unpatterned structures. However, in patterned structures the surface of implanted regions is found to be much rougher than unimplanted regions, and also rougher that implanted, unpatterned GaAs for the same annealing conditions. Atomic force microscopy study shows that the rms roughness is about 35 nm in the exposed regions. This indicates that materials sputtered away from the surface could be accumulated within the confined regions during the implant and this accumulation causes the rough surface. This roughness seems to have little effect on GaMn precipitate formation in implanted regions.

Since the inter-particle spacing in the implanted region is about 1 μm, it should be possible to fabricate one-dimensional lines of magnetic particles when the linewidth is further reduced to about 1 μm. Then, one can, in principle, use ion-implantation technique to make flexible patterned magnetic structures in GaAs semiconductors in a more controllable fashion.

In conclusion, we have successfully demonstrated that submicron room-temperature ferromagnets can be formed in GaAs by a simple process of ion implantation and subsequent heat treatment. Magnetic force microscopy has been shown to be a powerful tool for studying the magnetic state of submicron ferromagnetic structures and their magnetic anisotropy. With detailed knowledge of the magnetic state of MFM probes in changing fields, MFM can reveal a dynamic switching process of individual single-domain particles in an applied field. Such knowledge has been successfully obtained by doing MFM imaging on a miniature current-carrying strip. Furthermore, patterned ferromagnetic structures on GaAs have been fabricated using a SiO_2 mask during ion implantation. The method can be further refined to produce better controlled one-dimensional structures for other magneto-transport and optical studies.

ACKNOWLEDGMENTS

J. M. Kikkawa's assistance is greatly acknowledged. This work is supported by grants from the AFOSR, No. F49620-96-1-0118, and No. F49620-95-1-0434, and the NSF Center for Quantized Electronic Structures, No. DMR 91-20007.

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