Patterning of epitaxial MnAs on GaAs by direct optical writing

M. Ramsteiner,[a] H. J. Zhu, and L. Däweritz
Paul-Drude-Institut für Festkörperelektronik, Hausvogteiplatz 5–7, 10117 Berlin, Germany

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We demonstrate patterning of ferromagnetic MnAs layers on GaAs substrates by optical writing with a focused laser beam. Depending on the writing conditions, stripes with ridge- and groove-shaped cross sections can be produced without damage of the GaAs substrate. Using *in situ* control by resistance measurements, conditions for the preparation of completely insulating stripes can be established. The formation of insulating and paramagnetic Mn_3O_4 during optical writing is verified by Raman scattering. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1410356]

Epitaxial MnAs on GaAs belongs to a promising class of electronic material systems consisting of ferromagnetic metal(semi-metal)/semiconductor heterostructures, which are currently of great interest.[1–3] Potential device applications are nonvolatile memory and magnetic field sensors directly coupled with semiconductor circuits. For the fabrication of devices based on the hybrid material system MnAs on GaAs, it is indispensable to pattern the MnAs layers. Furthermore, patterning is a necessary tool for tailoring magnetic properties, i.e., to overcome the restrictions concerning the magnetization direction given by the shape anisotropy of extended thin films. The common method, photolithography in connection with wet/dry etching, is expensive and inconvenient. Focused laser beam writing, however, is a powerful and convenient alternative technique to pattern a variety of materials.[4,5] Through the use of holographic lithography, a standard method in semiconductor technology for the fabrication of lateral microstructures, direct optical writing allows for the periodic patterning of large areas.[6] In this letter, we demonstrate the direct patterning of epitaxial MnAs layers on GaAs by focused laser beams. Atomic force microscopy (AFM), Raman scattering, and photoluminescence (PL) spectroscopy have been used to characterize the patterned surfaces.

The MnAs layers were grown on semi-insulating GaAs(001) substrates at a temperature as low as 250 °C by solid-source molecular-beam epitaxy. The growth procedure is described in detail elsewhere.[3] MnAs layers of two different thicknesses, 20 and 270 nm, have been investigated. The optical writing was performed under ambient conditions. We used the 514.5 nm line of an Ar^+-ion laser focused by a microscope objective to a spot diameter of 1 μm on the MnAs surface. The laser spot was scanned along a line of adjustable length with a frequency of 20 Hz. The critical writing parameters are the laser power P_L, the scan length l_S, the scan duration t_S, and the MnAs layer thickness d_L. For micro-PL and Raman spectroscopy, the collected light was dispersed in a DILOR spectrograph XY800 and detected by a charge-coupled device array. The spectra were excited with the same laser line used for optical writing, but at reduced power. Micro-Raman line and area scans were performed with the samples mounted on a motorized XY translation stage.

The influence of the laser power on the optical writing efficiency exhibits a threshold-like behavior. For a thick MnAs layer and particular conditions ($d_L = 270$ nm, $l_S = 18$ μm, and $t_S = 60$ s), no material modifications can be achieved for $P_L < 15$ mW, even when t_S is strongly increased. For the same conditions, $P_L = 21$ mW leads to the formation of grooves induced by material evaporation, which are surrounded by convex regions. Furthermore, if t_S becomes too long, the good heat conduction of the relatively thick MnAs layer leads to surface modifications within 5 μm distance from the stripe center. For thin layers, choosing appropriate parameters, both narrow grooves and ridges can be obtained on the MnAs surface without modification of the surface at larger distances. AFM images of both kinds of stripes produced on the same 20 nm thick MnAs layer are shown in Fig. 1. The shape of the stripe cross section can be controlled by the writing power P_L. It should be emphasized that the power threshold in the writing efficiency decreases with d_L according to the smaller heat conduction of thin MnAs layers and the reduced volume heated by the absorbed laser light. The groove-shaped stripe for the thin MnAs layer ($d_L = 20$ nm) was obtained with $P_L = 11$ mW [Fig. 1(a)], whereas the ridge-shaped stripe [Fig. 1(b)] was realized by decreasing the power to $P_L = 8$ mW. As demonstrated by *in*

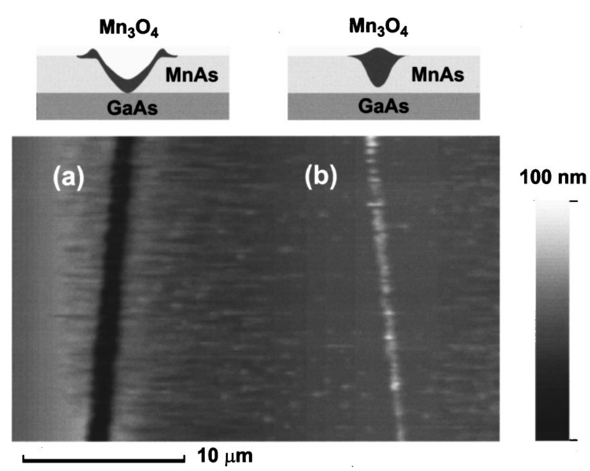

FIG. 1. AFM image of (a) a groove and (b) a ridge produced by optical writing on MnAs. The schematic diagrams on top display the corresponding cross sectional structures.

[a]Electronic mail: mer@pdi-berlin.de

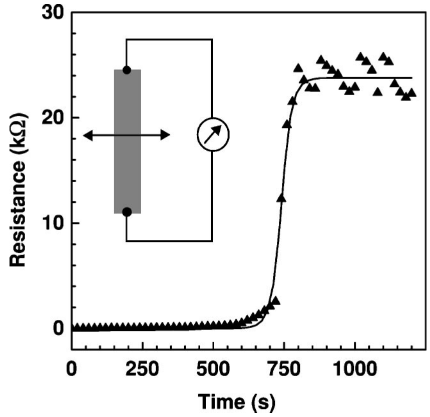

FIG. 2. Time dependence of resistance between two ends of a 28 μm wide MnAs bar separated by a focused laser scan for $d_L = 20$ nm and $P_L = 11$ mW. The diagram indicates the measurement configuration.

situ resistance measurements and optical spectroscopy, completely insulating and transparent (for visible light) stripes can be achieved for both types of cross sections.

The insulating property of the stripes has been demonstrated by dividing MnAs bars into two parts by optical writing. During the scanning of the focused laser across a 28 μm wide MnAs bar, the resistance was measured *in situ* between the two ends of the MnAs bar (cf. inset in Fig. 2). Following a constant low resistance of the metallic MnAs bar, the time-dependent resistance shown in Fig. 2 reveals a strong and steep increase after a certain scan duration, which strongly depends on the laser power. The data shown in Fig. 2 have been aquired by using a laser power close to the threshold for the optical writing efficiency in order to enlarge the time scale, on which the MnAs bar was devided into two parts. The strong resistance increase is followed by a saturation at about 25 kΩ. Switching the laser off leads to a subsequent increase of the resistance to about 20 MΩ. This result demonstrates the feasibility of optical writing to produce completely insulating stripes. The residual conductivity (resistance of 25 kΩ) during the laser scan is explained by the photoconductivity of the GaAs substrate.

For transparent stripes, micro-PL and Raman spectroscopy has been used to characterize the underlying substrate. Certain optical writing conditions lead to the diffusion of Mn into the GaAs substrate. Such a case is documented by the PL spectrum in Fig. 3(a), which reveals besides the near band gap recombination due to donor bound excitons and carbon acceptors (C_{As}) a peak at 1.41 eV caused by Mn_{Ga} acceptors in the GaAs substrate.[7] However, using appropriate conditions, it has been found that the influence of optical writing on the GaAs substrate properties is negligible.

A typical Raman spectrum taken in the vicinity of a transparent groove- or ridge-shaped stripe is shown in Fig. 3(b). The transparency, which indicates a complete removal of the opaque MnAs, is revealed by the observation of the longitudinal optical (LO)–phonon line from the GaAs substrate at 292 cm^{-1}. The narrow width of the LO–phonon line indicates that the optical writing can be performed without any damage to the GaAs substrate. More interestingly, the Raman peak at 660 cm^{-1} provides evidence for the formation of manganese oxide. This peak together with the weaker peaks at 375 and 320 cm^{-1} identifies the manganese oxide as

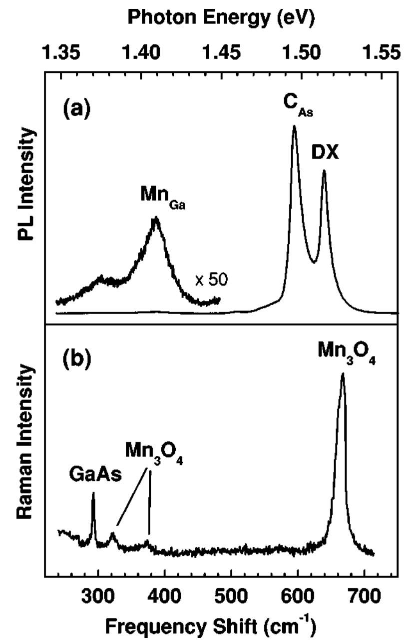

FIG. 3. (a) Low-temperature ($T = 10$ K) PL spectrum and (b) room-temperature Raman spectrum of a MnAs/GaAs sample in the vicinity of a focused laser scan.

the transparent, insulating and paramagnetic Mn_3O_4.[8] The spatial variation of the Mn_3O_4 formation as well as of the transparency has been measured by micro-Raman line and area scans. Figure 4(a) shows the intensity of the LO-phonon line from the GaAs substrate together with that of the Mn_3O_4 peak at 660 cm^{-1} as a function of position for a line scan across the stripes displayed in Fig. 1. The intensity maxima coincide at the positions of the groove- and ridge-shaped stripes, and the widths at half maximum are 1.3 and 0.8 μm

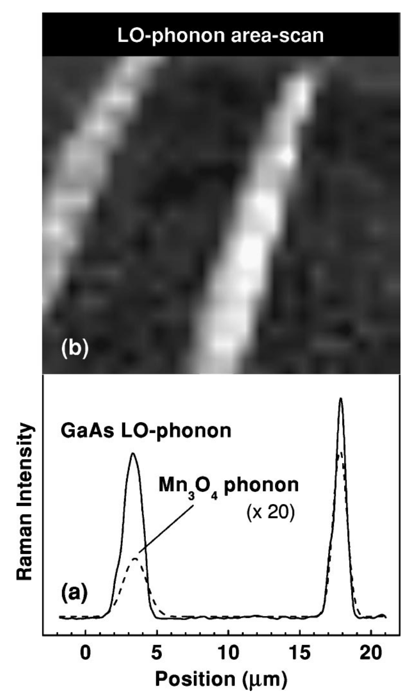

FIG. 4. (a) Raman intensities of GaAs (solid line) and Mn_3O_4 (dashed line) phonon lines as a function of position for a line-scan across the stripes displayed in Fig. 1. (b) Raman intensity of the GaAs LO-phonon line for a 25×25 μm^2 area scan across both stripes displayed in Fig. 1. Regions with large Raman intensities are indicated by bright areas.

for the GaAs signal. This result reveals that the complete removal of the opaque MnAs is restricted to stripes of only 1 μm width. Consequently, the 2–3 μm wide convex regions, which surround the groove-shaped stripes [cf. Fig. 1(a)], are most likely formed on top of the MnAs surface and do not affect the patterning resolution of about 1 μm. The Raman intensity of the GaAs LO-phonon line for a $25\times25\ \mu m^2$ area scan across the same stripes is shown in Fig. 4(b). The bright regions of high Raman intensities reveal the transparency of the whole stripes prepared by optical writing. It should be mentioned here that, using an appropriate GaAs/(Al,Ga)As heterostructure as a template for the MnAs growth, depth-resolved details of the optical writing influence can be gained by Raman spectroscopy. Using a simple layer sequence like MnAs/GaAs/(Al,Ga)As, it can be tested for different conditions as to whether or not there is an influence of the optical writing on the top GaAs layer or even on the (Al,Ga)As layer below.

On the basis of our topographic and spectroscopic investigations, we attribute the optical writing process to evaporation and oxidation due to absorption induced heating by the focused laser light (cf. schematic diagrams in Fig. 1). Groove-shaped stripes are formed by both evaporation and oxidation. The convex regions, which surround the groove-shaped stripes, are presumably formed by redeposition and oxidation of evaporated material. Ridge-shaped stripes are formed at lower P_L, where the temperature induced by the laser heating is high enough to promote oxidation (by breaking Mn–As bonds), but too low for evaporation. A critical temperature regime may be defined by the sublimation temperature of As at 614 °C, which could play a role in terms of chemical reactivity.[9] Both processes, the evaporation and the oxidation, lead to transparent, insulating and nonferromagnetic (Mn_3O_4 is paramagnetic) areas. Further work has to be done to study the electrical and magnetic properties of patterned MnAs regions, especially in the vicinity of the transparent stripes produced by optical writing.

In conclusion, we have demonstrated the feasibility of direct optical writing to pattern MnAs layers on GaAs by producing insulating and nonferromagnetic areas. Our results pave the way for large-area patterning of MnAs by holographic lithography. First arrays of below 1 μm wide stripes have already been produced.

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