

Atom lithography of Fe

E. te Sligte, B. Smeets, K. M. R. van der Stam, R. W. Herfst, P. van der Straten, H. C. W. Beijerinck, and K. A. H. van Leeuwen^{a)}
Eindhoven University of Technology, P.O. Box 513, 5600 MB Eindhoven, The Netherlands

(Received 9 July 2004; accepted 13 September 2004)

Direct write atom lithography is a technique in which nearly resonant light is used to pattern an atom beam. Nanostructures are formed when the patterned beam falls onto a substrate. We have applied this lithography scheme to a ferromagnetic element, using a 372 nm laser light standing wave to pattern a beam of iron atoms. In this proof-of-principle experiment, we have deposited a grid of 50-nm-wide lines 186 nm apart. These ultraregular, large-scale, ferromagnetic wire arrays may generate exciting new developments in the fields of spintronics and nanomagnetism. © 2004 American Institute of Physics. [DOI: 10.1063/1.1818347]

Atom lithography¹ is the logical complement of conventional lithography. Whereas conventional lithography uses mechanical masks (i.e., atoms) to impose a pattern onto a light beam, atom lithography uses light masks to transfer a pattern into an atom beam. The resolution of conventional lithography is limited by the wave character of the light used. Applying the same criterion to atom lithography, we find that the de Broglie wavelength of the atoms (typically 0.01 nm) is small compared to their size. Fully quantum mechanical calculations typically yield feature size limits below 10 nm.² This resolution, coupled with the fact that a light mask can be applied over as large an area as desired,³ makes atom lithography a useful lithographical technique.

The underlying principle of atom lithography is the interaction of a light field with the electrical dipole it induces in atoms. For a light mask to provide a large deflective force, its frequency must be chosen close to an atomic resonance. This technique was first applied to a Na beam,⁴ which is relatively easy to manipulate optically, but technologically of little value. A major step forward was the application of atom lithography to Cr.⁵ Since then, Al⁶ has been used, and experiments are under way on Si⁷ and various group III elements.⁸ The main limiting factor in the application of atom lithography to new materials is the availability of sufficiently stable and powerful continuous wave lasers at the required wavelengths.

This letter reports on the application of this technique to a ferromagnetic element.⁹ Regular arrays of ferromagnetic lines or dots are of considerable interest for nanomagnetic and spintronic research.¹⁰ The relevant optical properties of the three ferromagnetic transition metals are shown in Table I. As can be seen, the wavelengths of all transitions are in the near UV range. Due to the large number of hyperfine states, and the awkward wavelength of the transition, cobalt can be ruled out as a candidate element. Both Fe and Ni are suitable for atom lithography. We opt for using Fe in our experiment as its saturation magnetization is greater.

For optimal structure resolution, it is essential that Fe nanostructures be deposited using a well-collimated atom beam. This collimation can be achieved by aperturing, which would result in dramatic flux losses, or by laser cooling. Laser cooling¹² has the potential to dramatically increase the

brightness of an atomic beam, in addition to collimating it. It does, however, require atoms to scatter a large number of photons. In practice, this means that the cooling transition has to be closed. In the case of Fe, there is a small, but significant, leak in the 5D_4 – 5F_5 transition, limiting the laser cooling efficiency that can be achieved. Figure 1 shows the level scheme of Fe, including leak rates. As can be seen, the dominant leak has a wavelength of 501 nm. Closing only this leak would reduce the leak rate from 1:243 to 1:1398. We do not implement such a repumping scheme at present.

Our experimental setup is shown schematically in Fig. 2. Fe atoms from a source described elsewhere¹³ are laser cooled by nearly resonant laser light. The source was operated in effusive mode, using a 1 mm nozzle at 1920 K. The laser system (not shown) is based on a Ti:sapphire laser producing up to 2 W of light at 744 nm. This light is coupled into a ring cavity, where it is frequency doubled by a nonlinear LBO crystal. The frequency of the laser light is stabilized using a hollow cathode discharge.¹⁴ The atom beam that impinges on the sample typically has a divergence of 1.5 mrad half width at half maximum (HWHM). Laser cooling reduces this divergence to around 0.2 mrad HWHM, good enough for atom lithography. Due to the leak, the flux of atoms in the atomic ground state is only increased by a factor 1.8.

The collimated atom beam is subjected to the simplest light mask possible—a one-dimensional standing wave. The dipole potential generated by the standing light wave of intensity $I(\mathbf{r})$ acting on a two-level atom can be written as

TABLE I. The optical properties of the three ferromagnetic transition metals. Leak ratios determined from NIST Atomic Spectra Database (Ref. 11) where available.

Atomic species	⁵⁹ Co	⁵⁶ Fe	⁵⁸ Ni
Abundance	100%	92%	68%
Ground state	$a^4F_{9/2}$	a^5D_4	a^3F_4
Nuclear spin	7/2	0	0
Transition to	$x^4G_{11/2}^\circ$	$z^5F_5^\circ$	$z^3G_5^\circ$
Wavelength (nm)	240.5	372.0	323.4
Linewidth (MHz)	57	2.58	1.16
Saturation intensity (W/m ²)	5156	62	43
Leak ratio	...	1:243	...

^{a)}Electronic mail: k.a.h.v.leeuwen@tue.nl

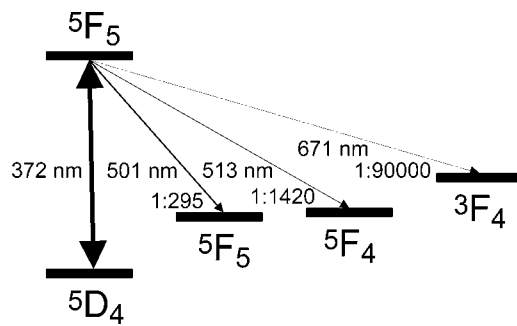


FIG. 1. Level scheme of Fe, showing the cooling transition and the three leaking transitions. The total leak rate is 1:243. A 501 nm repumper would reduce the leak by a factor 5.7.

$$U_{\text{dip}} = \pm \frac{\hbar}{2} \sqrt{\delta^2 + \Gamma^2} \frac{I(\mathbf{r})}{2I_s} - \frac{\hbar\delta}{2} \quad (1)$$

under the assumption that spontaneous emission does not occur.¹⁵ Here, Γ is the natural linewidth, δ the detuning, and I_s is the saturation intensity of the transition (Table I). The plus sign applies for blue detuning, the minus sign for red detuned standing waves. As our standing wave is a Gaussian beam that reflects off a dielectric mirror, we can use

$$I(\mathbf{r}) = \frac{8P}{\pi w^2} \sin^2(kx) \exp\left(-2\frac{y^2 + z^2}{w^2}\right), \quad (2)$$

with w the beam waist radius, and P the traveling wave power.

Assuming the atoms travel in the z direction, they “see” only one period of the standing wave. Taylor expansion of the potential around the minimum then yields a harmonic oscillator potential, which acts as a lens on the atoms. The atoms thus get focused toward the potential minima. For ease of alignment, it is preferable to focus the atoms in the center of the light mask as seen along the atoms’ propagation direction. The optimum laser beam power for a such a circular Gaussian lens array can be evaluated numerically for a two-level atom:¹⁶

$$P_f = 5.37 \frac{\pi E_{\text{kin}} I_s \delta}{\hbar \Gamma k^2}, \quad (3)$$

where E_{kin} is the kinetic energy of the atoms being focused, and k the wave number of the laser light. For Fe, after correction for the Zeeman sublevel structure of the transition, this power is ≈ 13 mW at a detuning of 150 MHz.

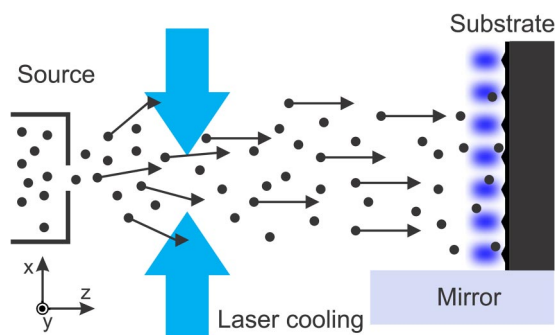


FIG. 2. Schematic of the experimental scheme. Fe atoms (circles) from source (left) are laser cooled (center) and then deposited through a light mask (right).

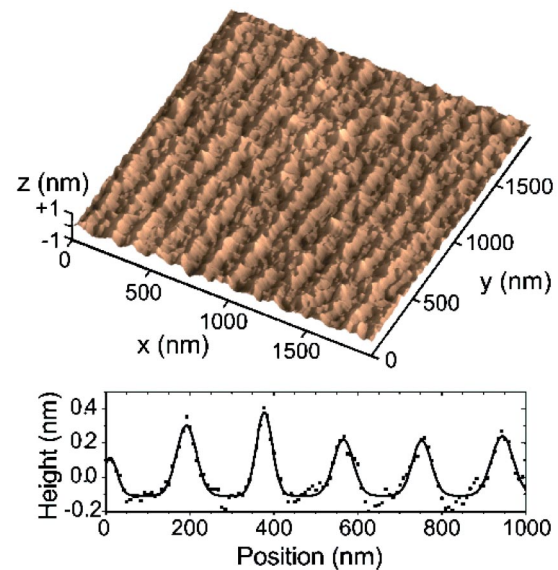


FIG. 3. Nanostructures from atomic Fe. Top: AFM scan of surface. Bottom: integrated line profile. Lines are approximately 50 nm wide and spaced at 186 nm intervals.

In our experiment, we use an AOM to detune part of the laser light by 150 MHz ($\approx 58 \Gamma$). The light mask is created by focusing a circular $w = 0.85 \pm 0.05$ mm laser beam using an antireflection coated lens of focal length 200 mm. Gaussian beam optics predicts a beam radius in the focus of 30 μm ; we measured $45 \pm 1 \mu\text{m}$ using a 10 μm slit. The running wave power in the light mask is 10 mW. All relevant angles in the experiment were aligned to within 0.5 mrad.

The sample holder is made of a single piece of stainless steel. Rigidly clamping both the substrate and the mirror that creates the light mask to this piece of stainless steel ensures optimum stability of the substrate with respect to the standing wave. The main source of variation in this alignment—and thus of blurring in the structures deposited—is thermal expansion of the substrate during deposition.¹⁷ Our sample holder design includes a Macor spacer block between the sample and the sample holder to provide electrical isolation, allowing the sample to be put on high voltage. This enables e-beam bakeout of conducting substrates. Presently, our substrate consists of a piece of monocrystalline Si[100] wafer with the native oxide layer still intact. We used ethanol to rinse the sample prior to installing it in the vacuum system, but applied no further cleaning. We do not actively stabilize the temperature of the sample.

An AFM scan of the nanostructures that were created in this initial deposition experiment is shown in Fig. 3. The period of the nanostructures was determined by comparison with nanostructures created using metastable He atom lithography,¹⁸ with a period of 542 nm. Using this reference sample, we found a period of 189 ± 4 nm, in agreement with the nominal value of 186.05 nm. On the same length scale, the width of the lines was 55 ± 5 nm. The height of the structures was relatively low, about 0.6 nm at an exposure to approximately 5 nm of Fe flux. We attribute this poor contrast ratio to three distinct effects. First, in the case of thermally evaporated Fe, only around 50% of the atoms is in the electronic ground state. All other atoms do not interact with any of the light fields involved. Second, laser cooling on the leaky transition causes the loss of a significant fraction of the

atoms, which also contributes to the high background flux. Finally, even in a perfectly collimated beam in which all atoms respond to the light field, there will be atoms that impinge well outside the potential minima, causing an additional background term that is always present in direct-write atom lithography. We hope to improve the beam flux greatly by installing a two-dimensional laser cooling section and by using a higher-temperature evaporation source. Given the relatively narrow structure widths obtained, we do not expect surface diffusion to play an important role in the poor contrast of the structures.

In conclusion, this letter reports on a periodic array of ferromagnetic Fe nanostructures using atom lithography, and demonstrates the applicability of the technique to magnetic materials. In the future, we will improve the quality of the nanostructure deposition process, and investigate their magnetic properties. We are presently working to increase the Fe flux of the source. Furthermore, an evaporation source has been installed that will allow us to cover the Fe nanostructures with a thin Ag film to prevent corrosion and oxidation, and the concurrent loss of magnetic order. We are also exploring two further options to improve the contrast ratio: first, using a repumper laser to reduce the influence of the leak; and second, transferring the atoms from the ground state into a low-lying metastable state from which a closed transition is available. Either option will drastically increase the usable flux of Fe atoms. The latter will also allow us to physically separate the atoms in the correct state from the background atoms.

This work is financially supported by the Dutch Foundation for Fundamental Research on Matter (FOM). The au-

thors would also like to thank the VU Amsterdam for the use of one of its samples.

¹D. Meschede and H. Metcalf, *J. Phys. D* **36**, R17 (2003).

²R. C. M. Bosch, Ph.D. thesis, TU/e, Eindhoven, 2002.

³R. E. Behringer, V. Natarajan, and G. Timp, *Appl. Phys. Lett.* **68**, 1034 (1995).

⁴G. Timp, R. E. Behringer, D. M. Tennant, J. E. Cunningham, M. Prentiss, and K. K. Berggren, *Phys. Rev. Lett.* **69**, 1636 (1992).

⁵J. J. McClelland, R. E. Scholten, E. C. Palm, and R. J. Celotta, *Science* **262**, 877 (1993).

⁶R. W. McGowan, D. Giltner, and S. A. Lee, *Opt. Lett.* **20**, 2535 (1995).

⁷H. Kumagai, Y. Asakawa, T. Iwane, K. Midorikawa, and M. Obara, *RIKEN Rev.* **50**, 34 (2003).

⁸S. J. Rehse, R. W. McGowan, and S. A. Lee, *Appl. Phys. B: Lasers Opt.* **70**, 657 (2000).

⁹While writing this letter, we learned that a group of Prof. Th. Rasing of the Radboud University of Nijmegen has achieved similar results in the same timeframe. G. Myszkiewicz, J. Hohlfeld, A. J. Toonen, A. F. Van Etteger, O. I. Shklyarevskii, W. L. Meerts, Th. Rasing, and E. Jurdik, *Appl. Phys. Lett.* **85**, 3842 (2004).

¹⁰*Nanostructured Magnetic Materials and their Applications* edited by D. Shi, B. Aktaş, L. Pust, and F. Mikailov (Springer, Berlin, 2002).

¹¹http://www.physics.nist.gov/cgi-bin/AtData/main_asd.

¹²H. J. Metcalf and P. van der Straten, *Laser Cooling and Trapping* (Springer, Berlin, 1999).

¹³R. C. M. Bosch, H. C. W. Beijerinck, P. van der Straten, and K. A. H. van Leeuwen, *Eur. Phys. J.: Appl. Phys.* **18**, 221 (2002).

¹⁴B. Smeets, R. C. M. Bosch, P. van der Straten, E. te Sligte, R. E. Scholten, H. C. W. Beijerinck, and K. A. H. van Leeuwen, *Appl. Phys. B: Lasers Opt.* **76**, 815 (2003).

¹⁵J. Dalibard and C. Cohen-Tannoudji, *J. Opt. Soc. Am. B* **2**, 1707 (1985).

¹⁶J. J. McClelland, *J. Opt. Soc. Am. B* **12**, 1761 (1995).

¹⁷J. J. McClelland, W. R. Anderson, C. C. Bradley, M. Walkiewicz, R. J. Celotta, E. Jurdik, and R. D. Deslattes, *J. Res. Natl. Inst. Stand. Technol.* **108**, 99 (2003).

¹⁸S. J. H. Petra, L. Feenstra, W. Hogervorst, and W. Vassen, *Appl. Phys. B: Lasers Opt.* **78**, 133 (2004).