Nanostructure fabrication via laser-focused atomic deposition (invited)

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Nanostructured materials and devices will play an important role in a variety of future technologies, including magnetics. We describe a method for nanostructure fabrication based on the use of laser light to focus neutral atoms. The method uses neither a mask nor a resist, but relies on the direct deposition of atoms to form permanent structures. Since the atomic de Broglie wavelength is of picometer order, the size of structures produced is not significantly limited by diffraction, as in optical lithography. Lines as narrow as 38 nm full width at half maximum spaced by 213 nm have been produced and we have demonstrated the production of a two-dimensional array of dots. The highly parallel process of nanostructure formation and the intrinsic accuracy of the optical wavelength that determines structure spacing suggest a number of interesting applications, including calibration standards for various types of microscopy, lithography, and micromeasurement systems. Possible magnetic applications include the production of arrays of magnetic elements, laterally structured giant magnetoresistive devices, and the patterning of magnetic media.
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I. INTRODUCTION

The fabrication and use of nanostructures, both as tools to further our understanding of physical principles and to provide novel functionality in devices, continues to expand unabated. Nanostructures are generally understood to possess at least one dimension on the nanometer scale, more typically below 100 nm. Frequently, the near atomic scale of the dimension gives rise to unique properties to be explored or exploited. While this definition includes the area of thin-film research, such work is usually not discussed as nanostructure science. However, multilayer structures of thin films are included within the definition. This general area of research has also been referred to as nanotechnology.

While far more work has been performed on the electronic characteristics of small structures, a growing body of research concerns the fabrication and properties of magnetic nanostructures. A great deal of current research is focused on giant magnetoresistive (GMR) devices generally involving multilayer structures.[1] One recent article reports the fabrication of a GMR device consisting of a series of GMR multilayer structures with all three dimensions in the nanometer range.[2] Another reports the use of a wedge-shaped nanostructure to systematically study exchange-coupled magnetic layers.[3] Others concentrate on fabricating arrays of isolated magnetic elements to study their interactions,[4–7] magnetic properties,[8,9] or magnetic quantum tunneling.[10] Applications include improved MFM tips[11] and high-density magnetic storage.[12]

The production of one-dimensional nanostructures is generally done with molecular beam epitaxy and/or sputter deposition. Structures with two and three dimensions in the nanometer range require a patterning technique, such as optical or electron-beam lithography. Optical lithography, while convenient because of its wide usage and ability to pattern large areas in parallel, is limited because of the diffraction of light to feature sizes of about 180 nm, i.e., just at the upper limit of the nanoscale regime. As a result, electron-beam lithography, which has an insignificant diffraction limit, has generally been the method of choice for fabricating higher-dimensional nanostructures. Features as small as 20 nm can be fabricated with relative ease, and in some special cases, features as small as 1–2 nm have been achieved.[13] Further progress has been made recently by replacing the electron beam with a scanning tunneling microscopy (STM) tip.[14] Despite its popularity and potential for high resolution, there are drawbacks associated with electron-beam lithography. Because patterns are formed by scanning a finely focused beam, structures are generated serially across a substrate. There is also an inherent trade-off between speed and resolution. Since electrons mutually repel each other, high resolution can only be achieved at the expense of beam current. Thus the beam must be scanned relatively slowly across the surface in order to fully expose the resist. Besides introducing problems like sample drift, proximity effects, and stitching errors, this limits the complexity of the pattern to be fabricated: a large array of very small features can require an exposure of many hours, a length of time that rapidly becomes impractical.

II. LASER FOCUSING OF ATOMS

Neutral atom focusing is a completely different approach to nanostructure fabrication. While still in its infancy, it already addresses several of the stumbling blocks of present methods. The diffraction limit is insignificant, as it is for electron beams, since the de Broglie wavelength of the atom is typically in the picometer range. Unlike electrons, however, the atoms are electrically neutral, so there is no space-charge repulsion limiting the flux in finely focused regions. Furthermore, the focusing scheme can be made massively parallel, allowing large areas to be patterned rapidly. In addition, the focusing schemes generally make use of highly stable optical wavelengths, so highly accurate patterns can be produced over relatively large areas without stitching errors. Still another advantage is that the process can be implemented in a direct-write manner, eliminating the need for a

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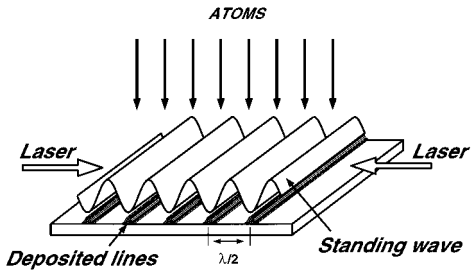

FIG. 1. Schematic of laser-focused atomic deposition process.

physical mask and subsequent processing. Though, so far, no direct application to the fabrication of magnetic nanostructures has been carried out, it appears that many of these advantages could prove useful for such applications.

The focusing of neutral atoms is achieved by configuring electromagnetic fields in such a way as to exert forces on neutral atoms toward an axis. The study of the motion of atoms in electromagnetic fields is a major component of the new field of atom optics,[15] which treats the manipulation of atoms by devices that serve as lenses, mirrors, beamsplitters, and gratings in analogy with light optics. To date, most of the configurations used for focusing atoms have involved the use of laser fields in which the frequency of the laser is tuned very close to an atomic resonance. Some work has been done, however, on the focusing of atoms in a hexapole magnetic field.[16] Laser fields provide an especially good medium for generating the necessary fields for high-resolution focusing of atoms because gradients can be generated over optical-wavelength distances, and tuning near resonance can greatly enhance the interaction. In particular, making use of a laser standing wave allows for massively parallel focusing of atoms in an array of lenses that is spaced with interferometric precision across a substrate (see Fig. 1).

In the presence of a near-resonant laser field, two types of radiation forces are present, the spontaneous force and the dipole force. The spontaneous force is simply the light pressure, i.e., the transfer of momentum that results when an atom absorbs a photon from the direction of the light source and reradiates a photon in an arbitrary direction. After a number of such interactions a beam of neutral atoms will be deflected away from the light source. The dipole force can be thought of as resulting from the interaction between an oscillating electric dipole induced in the atom and any spatial gradient that might be present in the oscillating laser electric field. Over the past 15 years, methods have evolved to use these forces, separately or in combination, to cool and trap,[17] and also focus,[18] atom beams.

The first application of laser focusing of atoms to controlled deposition involved observing the ''shadow'' of a resonant laser beam traversing a beam of sodium atoms depositing onto a surface.[19] This was followed by observation of optical diffraction from a grating of sodium atoms generated by focusing the atoms with the dipole force generated in the nodes of a standing wave passing across the surface of a substrate.[20] These experiments demonstrated the essential principle of laser focused atomic deposition but, because the

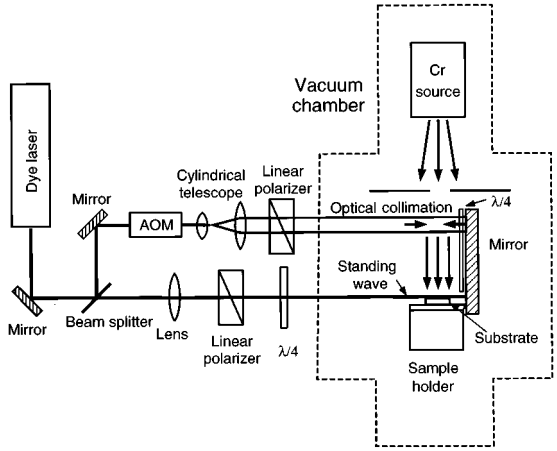

FIG. 2. Schematic of laser-focused atomic deposition apparatus, showing dye laser, acousto-optic modulator (AOM), miscellaneous optics (including quarter-wave plates, denoted by λ/4), vacuum chamber, Cr source, deposition substrate, and sample holder.

deposited atom was sodium, it was difficult to proceed with nanostructure fabrication, or carry out quantitative studies of the process. Sodium was chosen because it was easy to make an atomic beam and easy to tune a laser near the strong resonance at 589 nm. The first permanent laser-focused structures were fabricated using a chromium atomic beam, taking advantage of the atomic resonance at 425 nm.[21] This work has been followed recently by creation of structures in aluminum.[22] Because the use of a hard, fine-grained material such as chromium opens the possibility for precise fabrication and perhaps transfer of the patterns to other materials, and because of the relevance of chromium to magnetic materials, we discuss the results of the chromium research in some detail.

III. CHROMIUM EXPERIMENT

A schematic of the experimental arrangement[21] is shown in Fig. 2. In this experiment, Cr atoms effuse from a molecular beam epitaxy (MBE)-type evaporator, modified to produce a point source of atoms. They pass through a region where the beam is collimated optically, and then through a second, standing wave region where they are focused to form a pattern on a Si substrate. The optical standing wave, positioned immediately above the Si substrate, is formed by the reflection of a laser beam from a mirror in direct contact with the substrate. The laser wavelength used is 425.55 nm (in vacuum), which corresponds to the energy difference between the Cr $^7S_3$ ground state and the $^7P_4$ excited state, and the spacing between nodes of the resulting standing wave is just half that wavelength, or 212.78 nm. If the correct combination of laser beam size, intensity, wavelength, and position above the substrate is used, the standing wave acts as an array of cylindrical lenses to focus the atoms to form lines on the substrate.[21]

The dipole force is not strong, so the lenses described above are best thought of as weak immersion lenses. In order for their effect to be enough to bring the atoms into a fine focus, the incident atom beam must be highly collimated,

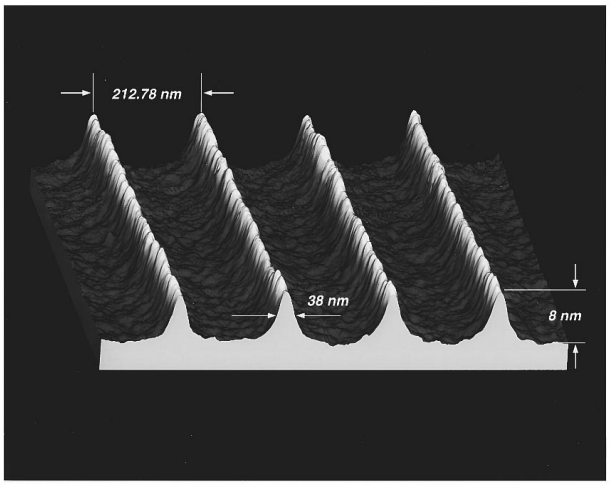

FIG. 3. Atomic force microscope image of Cr lines formed by laser-focused atomic deposition. The lines in this image have a height of 8 ± 1 nm.

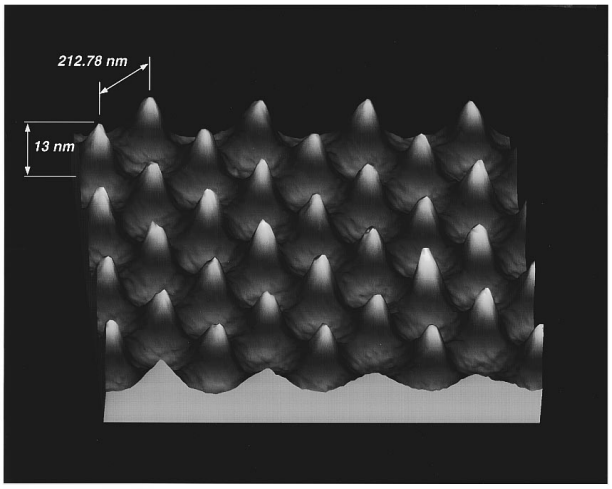

FIG. 4. Atomic force microscope image of a two-dimensional array formed by laser-focused atomic deposition of Cr.

possessing little transverse velocity. To achieve this, a collimation region is used, in which counterpropagating laser beams transverse to the atom beam are tuned to a frequency just below that of the resonant transition. In this process, photons are absorbed only if the transverse velocity of the atoms is large enough to Doppler shift the photon into resonance with the atom. In that case, the atoms will experience a momentum change that moves them closer to an ideally collimated beam. This collimation or laser cooling process, which is sometimes referred to as ''optical molasses,'' has been studied in great depth in one, two, and three dimensions in relation to laser cooling and trapping of atoms.[23] Using a variant of the basic process, which makes use of polarization gradients in the laser beams,[24] an atom beam collimation of the order of one part in 7000 is achieved.

IV. CHROMIUM RESULTS

Using the apparatus schematically shown in Fig. 2, samples were fabricated with Cr lines covering an area of up to 1 mm$\times$0.40 mm. Figure 3 shows an atomic force microscope (AFM) topograph of one of these samples. The lines are spaced by half the laser wavelength, 212.78 nm. The average height of the lines in Fig. 3 is 8 ± 1 nm,[25] and the full width at half maximum (FWHM) is 38 ± 1 nm, uncorrected for AFM tip shape (which could have an influence in this size range). The evaporation time, using a modest Cr oven, was 10 min. Although the AFM cannot distinguish between bare Si and Cr, we infer that there is a thickness of Cr in the valleys between the lines in this sample, based on measurements of the total atomic flux. We estimate this background to be 10 ± 4 nm thick. A complete understanding of the background is still to be established, however significant contributions include 16% of other isotopes of Cr that do not interact with the laser, about 10% Cr atoms that decay into a metastable ^5D level and no longer interact with the laser, and a possible high-velocity tail in the laser-cooled transverse velocity distribution.

In addition to the one-dimensional array of lines shown in Fig. 3, a two-dimensional array has also been created.[26] Figure 4 shows an AFM topograph of a section of a two-dimensional array of Cr ''dots'' made by first optically collimating in two dimensions and then using two standing waves positioned at right angles to each other. These dots cover an area of approximately 100 μm$\times$200 μm on the sample, have a FWHM of 80 ± 10 nm, and have a height of 13 ± 1 nm.

While the extension of the laser focusing process to two dimensions may seem straightforward, there are some subtleties that must be understood. When two standing waves that are temporally coherent (e.g., originating from the same laser) are superimposed upon each other at right angles, the resulting intensity pattern will, in general, depend on the relative temporal phase. However, for the special situation of orthogonal linear polarizations (one parallel to the substrate, the other perpendicular), this dependence is eliminated, allowing the laser focusing to be carried out without stabilizing the relative phase of the standing waves.

V. METASTABLE RARE GASES

Although the bulk of the work on laser focusing of atoms has been done with ''direct-write'' materials such as sodium, chromium, and aluminum, a new possibility has been suggested that makes use of metastable rare gases to exposure a lithographic resist.[27] The potential for this approach lies in the combination of lithography's ability to work with a wide range of materials with the atom optics advantages of high resolution, parallel fabrication, and low substrate damage. Metastable rare gases are both accessible to laser manipulation techniques, and they also have internal energy of up to 20 eV, sufficient to chemically alter a resist, yet not damage a substrate.

The ability of metastable atoms to act as a writing tool on a resist has been demonstrated using self-assembled monolayers (SAMs) of alkanethiolates on gold-coated silicon.[27] The SAM coating acts as a very thin resist, preventing etching of the gold when it is placed in an aqueous

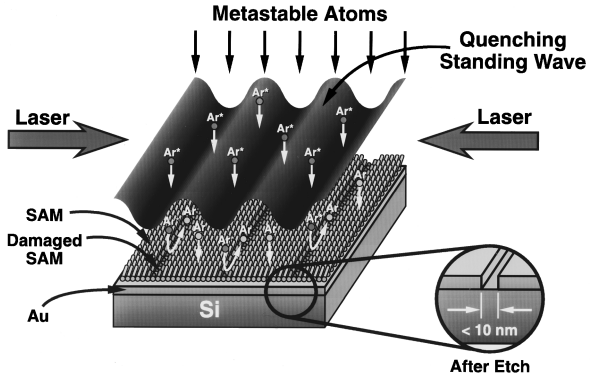

FIG. 5. Schematic of metastable rare gas lithography concept.

solution of ferricyanide. In regions where metastables have been allowed to strike the surface, the SAM is damaged, allowing the etch to penetrate.

So far, an upper bound of around 100 nm has been set for the resolution of this process by examining the edge roughness of a physical mask. The methods of laser manipulation have yet to be applied; however, work is ongoing, and several possibilities exist, using either 1.083 μm light on the He $(2^3S \rightarrow 2^3P)$ transition or 811 nm light on the Ar $(1s_5 \rightarrow 2p_9)$ transition. Metastable Ar opens a particularly interesting additional possibility in that it is possible to quench the metastable state using laser light at 764 or 801 nm. This allows the spatially selective removal of metastable atoms from the beam as an alternative to focusing (see Fig. 5), suggesting the possibility of significantly less background exposure. Preliminary estimates indicate that this quenching approach may result in features as small as 10 nm, though more work needs to be done to investigate this.

VI. FUTURE RESEARCH

We have only begun to investigate the application of atom optics to the manipulation of atoms as they impact a surface. There are a large number of possibilities that could lead to exciting new fabrication techniques. Given the current circumstances, a few areas for future improvement can be discussed, though many more may become apparent as the field develops.

Feature size is one area where improvement can be anticipated. Currently features as small as 38 nm FWHM have been created, but calculations of the focusing process, both semiclassical[28] and fully quantum,[29] predict that features as small as 10 nm should be attainable. This reduction in feature size will most probably arise from an improvement in the atomic source: reduction of the velocity spread, and still higher levels of collimation, appear to be the improvements that will help the most in this case.

The ability to create more complex patterns is also highly desirable if the process is to become useful for general nanostructure fabrication. To this end, arrays of complex patterns can be created by using the current two-dimensional process, perhaps with higher resolution, and simply scanning the substrate during deposition. In this way patterns can be ''painted'' within the unit cell of the standing wave, and these patterns will be repeated with very high accuracy across the substrate. Still more general patterns could be created by designing a more complicated optical field that puts the atoms exactly where they are required.

Another avenue for exploration is the possibility of extending the process to other atomic species. While nanostructures of other materials can be produced by using the metastable rare gas process discussed above or using Cr as a mask for reactive ion etching, it is also of interest to ask what other materials could be used directly. The essential characteristics are that the atomic species be relatively easily evaporated, and that there be a resonance transition accessible to an available laser wavelength. The resonance transition must have as its lower state one that has a significant population of atoms, either naturally or prepared (as in the case of metastable atoms). The full range of materials has not yet been explored, however it appears that many of the metallic species that might be of interest have resonant transitions in the ultraviolet, in the range from 200 to 300 nm. At present these wavelengths are difficult, though not impossible, to access with a laser. Nevertheless, laser technology is improving rapidly, and it is likely that in the future these atoms will become accessible.

The application of these new techniques to the fabrication of magnetic nanostructures remains in the realm of the future. However, the unique capabilities inherent in these new techniques offer several advantages when compared with other lithographic methods, and the length scale of their applicability is an interesting one from a magnetic perspective. We plan to explore the use of this method for magnetic nanostructure fabrication in the near future.

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