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Quasiperiodic structures via atom-optical nanofabrication

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We deposit a laser-collimated chromium beam onto a substrate through a quasiperiodic laser standing-wave (SW) tuned above the atomic resonance at the ^{52}Cr transition $^7S_3 \rightarrow {}^7P_4^o$ at 425.55 nm. This SW is created by interference of five laser beams crossing in one point at mutual angles of 72°. The resulting chromium pattern on the substrate surface mimics the geometry of the SW and it is thus itself quasiperiodic. On a surface area of 0.2×0.2 mm^2 the spatial Fourier spectrum of the measured patterns is decagonal. Besides being of fundamental interest, this quasiperiodic nanofabrication via atom optics can find its applications in photonics.

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In the past decade there has been a great deal of research devoted to photonic crystals, mainly owing to their potential applications in information and communication technology. The structure of these materials is characterized by a high degree of order (periodicity or quasiperiodicity) and is designed such as to manipulate the propagation of light in a desired way. The most common approach to the fabrication of photonic crystals relies on a periodic modulation of the refractive index with periods on the order of the wavelength of light. In this scenario a complete (non-directional and for any polarization) photonic band gap (PBG) can be achieved only when the contrast of the refractive index becomes large. Consequently, the number of materials (from which a complete PBG can be made) is limited to those with a large refractive index, resulting in reduced optical transparency of devices. It was demonstrated that another approach, which is based on quasiperiodic ordering of photonic crystals, can overcome these difficulties as it allows for fabrication of structures with a complete PBG even from materials with low refractive index.[1] It is this latter concept to fabrication of photonic crystals that motivated us to carry out atom-optical deposition of quasiperiodic structures.

Atom optics—a technique to manipulate atomic trajectories by the use of electro-magnetic fields—can be applied to patterning of substrates or doping of materials with submicron structures. The main advantages of atom-optical fabrication are its compatibility with molecular beam epitaxy (i.e., the structure growth takes place *in vacuo* with no need for additional physical or chemical processing), its massiveness (i.e., a large surface area, in principle limited by the size of the atomic beam only, is patterned during one deposition run) and the unprecedented coherence of the deposition process.[2,3] To date, one of the most efficient methods applied to atom-optical structuring is laser-focused atomic deposition. This approach relies on interaction of atoms, prior to their deposition onto a substrate, with a far-off-resonant laser beam that exhibits gradients in the light intensity. In order to obtain gradients on a sub-wavelength scale, two or more laser beams can be allowed to interfere and thus to create a laser standing-wave (SW). Atoms, when they cross the laser SW, experience a spatially varying optical potential that, in the first approximation, follows the SW intensity profile.[4] When the laser frequency is tuned above an atomic resonance, the atomic beam is focused in each of the nodes of the SW. In contrast, negative detunings cause the atoms to gather in the regions of high light intensities. A substrate is then placed into the modulated atomic beam and sub-wavelength structures are grown.

Manipulation of atoms in a laser SW and subsequent fabrication of nanostructures was experimentally demonstrated in the direct deposition regime with atomic beams of sodium,[5] chromium,[6] and aluminum,[7] and via lithography with a self-assembled monolayer as the resist with cesium.[8] A number of different SW geometries were used: a one-dimensional SW created from two counter-propagating laser beams resulted in periodic nanolines;[5–8] a SW composed of four laser beams at right angles resulted in a square lattice of nanostructures;[9] and a SW obtained by three laser beams crossing at mutual angles of 120° resulted in a hexagonal nanostructured array.[10] More complicated periodic patterns were written by using more complex laser fields, such as those obtained by making use of polarization gradients,[11,12] by reflecting a laser beam from a holographic mirror,[13] by utilizing a slight misalignment of four initially orthogonal laser beams[14] or by beating of two atomic resonances.[3] All nanostructures fabricated via atom optics so far exhibited ''allowed'' symmetries, videlicet two-, four-, and six-fold symmetries.[15]

In this paper, we extend the technique of laser-focused atomic deposition to a quasiperiodic geometry. Five laser beams crossing at mutual angles of 72° are employed to create the SW. A chromium atomic beam is used and the SW laser frequency is tuned above the ^{52}Cr transition $^7S_3 \rightarrow {}^7P_4^o$ at 425.55 nm. The optical potential exhibits a ''forbidden'' ten-fold symmetry as does also the surface structure. The absence of translational symmetry and a non-crystallographic orientational order, that is associated with the decagonal symmetry axis, result in a structure with striking geometry and with potentially intriguing optical properties.

Prior to carrying out the experiment, we performed numerical calculations of the atomic flux distribution in a quasiperiodic optical potential. In Fig. 1(a) the optical intensity, $I(\mathbf{r})$, at the center of a Gaussian laser SW is shown. In the first approximation, the optical potential, $U(\mathbf{r})$, is proportional to $I(\mathbf{r})$,[4]

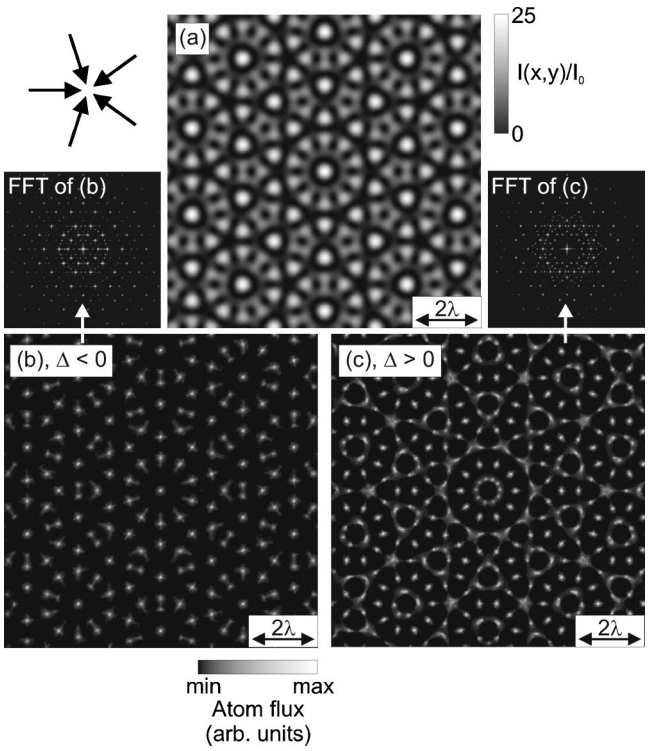

FIG. 1. Calculation of laser-focused deposition of a chromium beam. (a) Quasiperiodic SW intensity resulting from interference of five laser beams crossing at mutual angles of 72°. (b), (c) Atomic flux distributions for $\Delta<0$ (b) and $\Delta>0$ (c). Spatial Fourier spectra of (b) and (c) are also shown. Calculation parameters: $P=90$ mW, $W_0=200$ μm, $|\Delta|/2\pi=200$ MHz. 10^8 atomic trajectories were traced to obtain these results. For more details, see the text.

$$U(\mathbf{r})=\frac{\hbar\Gamma^2}{8\Delta}\frac{I(\mathbf{r})}{I_S}. \tag{1}$$

Here, $\hbar$ is Planck's constant divided by 2π, $\Gamma/2\pi=5$ MHz is the natural linewidth of the $^7S_3\rightarrow{}^7P_4^o$ ^{52}Cr atomic line, Δ is the detuning of the laser frequency from the atomic resonance, and $I_S=85$ W/m^2 is the atomic saturation intensity. Using this potential, the Newton equation of motion was integrated for 10^8 random atoms with initial conditions corresponding to the Maxwell-Boltzmann statistics of a thermal chromium beam that is collimated in the transverse directions to 0.3 mrad at full-width at half maximum (FWHM). The ''oven'' temperature was adjusted to 1900 K. The single-wave, traveling laser power was set to 90 mW. The five laser beams, that create the SW, were all focused down to a $1/e^2$ radius of 200 μm. The ground-state magnetic sublevel structure of ^{52}Cr as well as the presence of other chromium isotopes were taken into account.[16] The atomic flux distributions calculated at the center of the SW are shown in Figs. 1(b) and 1(c) for $\Delta/2\pi=-200$ and 200 MHz, respectively. The corresponding spatial Fourier spectra (or, equivalently, the far-field optical diffraction patterns) are also presented in Fig. 1. They clearly exhibit a ten-fold symmetry. We must note here that an experimental structure can differ from these calculations, because in reality it is extremely challenging to obtain a SW profile as that from Fig. 1. This is due to the physics of Gaussian laser beams as well as due to unavoidable diffraction on the substrate surface and possible phase fluctuations of the five laser beams. One can, however, approach such an ideal situation by properly tweaking alignment of the experiment. Then, the decagonal symmetry of the structure should become apparent, at least from the spatial Fourier spectra.

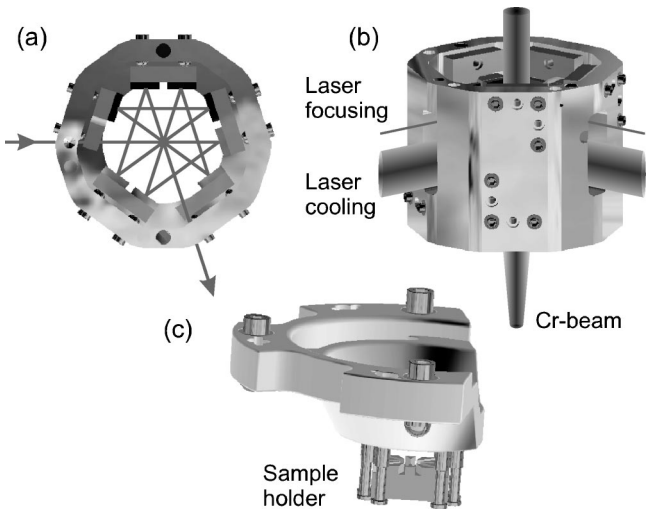

FIG. 2. *In vacuo* optical setup for fabrication of laser-focused quasiperiodic structures: (a) top view, (b) isometric view, (c) sample holder. The sample holder is placed on top of the unit from (b).

For our experiment, laser light at 425.55 nm was obtained by frequency doubling the output of a titanium-doped sapphire laser in an external enhancement doubling cavity.[17] A thermal beam of neutral chromium atoms was produced from a high-temperature effusion cell held at 1900 K. The vacuum pressure during the deposition was below 10^{-8} Pa. After passing a defining diaphragm (1.5×1.5 mm^2), the atomic beam was transversally collimated by means of laser cooling to a FWHM divergence angle of 0.3 ± 0.1 mrad.[18] Then, it was deposited onto a glass-ceramic substrate (rms surface roughness <5 Å) through a quasiperiodic laser SW. The laser cooling beam and the SW were respectively tuned 10 MHz below and 200 MHz above the involved chromium resonance. In order to assure mutual phase stability of the five laser beams creating the SW as well as respective alignment between the SW and the laser cooling beam, we designed a monolithic *in vacuo* unit made of high-strength aluminum (Fig. 2). Onto this unit eight dielectric mirrors (8 mm wide, 40 mm high) were mounted. These mirrors served to create both the laser molasses and the SW interference pattern. The optical path of the SW laser beam inside the unit is 466 mm. For this long distance, the $1/e^2$ laser beam radius, W_0, should not vary significantly in order to create an optical intensity distribution similar to that from Fig. 1(a). In contrast, tight focusing of the SW laser beam is required for high resolution, high contrast fabrication. Both these criteria cannot be realized simultaneously and thus a compromise must be reached. Therefore, we used a spherical lens with a long focal length of 2 m.[19] The SW laser beam was then focused to $W_0\simeq200$ μm and its confocal distance was $\simeq340$ mm. It

contained a power of 90 mW and was *s*-polarized. Using a highly accurate, specially fabricated alignment unit, the mutual angles of 72° of the five laser beams were adjusted to within 0.1 mrad. The substrate rested on a glass oblong (cut from a prism with optical quality faces) with grooves for the SW. This oblong was mounted into a holder that fits onto the main monolithic block (Fig. 2). The substrate was placed above the center of the SW beam such that only 30% of the total laser power was cut. This arrangement minimizes the diffraction problems and, following atom-optical calculations, still results in a higher density of the atoms near the SW nodes due to the effect of channeling.[16] The deposition time was set to 30 min, corresponding to an average thickness of the chromium film of ~ 10 nm.

We investigated the fabricated sample by means of atomic force microscopy (AFM) *ex vacuo*. A surface area of roughly 1.0×1.0 mm^2 exhibited modulation due to the presence of the SW during the deposition. Because at different positions the local intensities of the five laser beams differed, the SW pattern changed and so did also the sample morphology. For example, at those places where two of the five beams were dominant, the resulting structure consisted of nanolines with only a very slight modulation superimposed over them (due to the presence of the other three laser beams with low local intensities). At other places, more than two laser beams dominated and thus the modulation in both spatial directions became more pronounced. A more dot-like structure was grown. Spatial Fourier spectra of all these patterns showed a few very pronounced peaks positioned on circles. The angle spanned by any two of the peaks on the same circle was always an integer multiple of 36° to within a measurement uncertainty of 1.0°. When the five local intensities were comparable, the surface symmetry and the corresponding Fourier spectrum became almost decagonal. The term ''almost decagonal'' refers here to the fact that not all the Fourier peaks are equally intense. These decagonal structures covered a surface area of 0.2×0.2 mm^2. In Fig. 3(a) an AFM scan of such a structure is shown. From this raw AFM image with no pronounced modulation, the ten-fold symmetry is not immediately apparent. However, a spatial Fourier spectrum calculated from this image, shown in Fig. 3(a′), reveals ten clear peaks with different intensities on an inner circle and eight other peaks (plus additional two peaks with very low intensities) on an outer circle. The radii of these two circles correspond to wave vectors with magnitudes of $16.82\pm 0.31\ \mu\text{m}^{-1}$ and $27.18\pm 0.31\ \mu\text{m}^{-1}$, respectively. The ratio of these magnitudes evaluates to 1.614 ± 0.090, well agreeing with the golden ratio of the Fibonnaci sequence of $\simeq 1.618$. In Fig. 3(b′) a spatial Fourier spectrum ''cleaned'' from low (background, slow modulation) and high (noise) frequencies, employing a band-pass filter, is presented. An inverse spatial Fourier transform of this last spectrum shows a real space image of the quasiperiodic pattern on the substrate [Fig. 3(b)]. We note that the amplitude of the quasiperiodic surface modulation did not exceed 1.2 nm, corresponding to less than 5 monolayers of chromium.

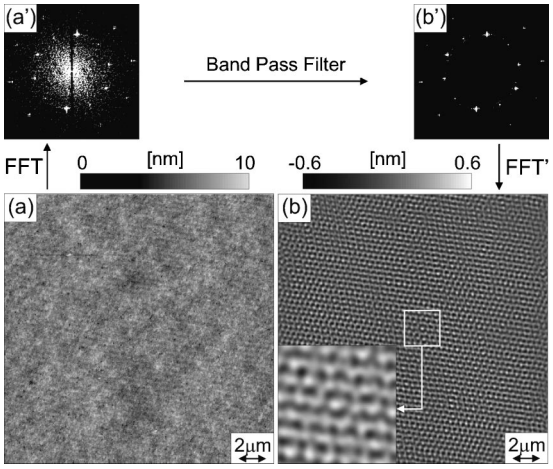

FIG. 3. Quasiperiodic chromium structures formed by laser-focused atomic deposition. (a) AFM scan of $20\times 20\ \mu\text{m}^2$ surface area. (a′) Spatial Fourier spectrum of the structure from (a). (b′) Filtered spatial Fourier spectrum of (a′)—bandpass filter. (b) Inverse spatial Fourier spectrum of (b′)—quasiperiodic surface modulation. Inset in (b): Zoom-in view of $2.5\times 2.5\ \mu\text{m}^2$ of the surface area enclosed in the white square.

In conclusion, we have demonstrated fabrication of quasiperiodic chromium structures on a sub-wavelength scale by means of laser-focused atomic deposition. Our first experiment shows that this atom-optical technique can be mastered to pattern a fairly large surface area with intriguing features that exhibit non-crystallographic symmetry. Although the contrast and the resolution of the sample fabricated by us remained limited, mainly owing to current experimental difficulties, the trend toward a decagonal symmetry is undisputable. Further technical improvements of our *in vacuo* optics will result in a better control of the SW interference pattern – a sharp focus and equal intensities of the laser beams that create it – allowing us to manipulate more efficiently the atomic trajectories in a quasiperiodic optical potential. Then, the result will be a high quality nanostructured array with the desired ten-fold symmetry. The fact that atom-optical deposition is compatible with molecular beam epitaxy, combined with the extreme coherence of the fabrication process, makes it an attractive tool for fabrication of structures for photonic manipulation of light. By this work we added a new dimension to this technique, thereby extending the gallery of its possible applications.

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[18] All quoted uncertainties are intended to be interpreted as one standard deviation combined random and systematic errors.
[19] Alternatively, the SW laser beam could have been refocused within the *in vacuo* optical unit. This solution resulted in a considerably more difficult alignment procedure and did not prove to be useful for a first demonstrative set of experiments.