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Atomic Nanofabrication: Perspectives for serial and parallel deposition

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Abstract. Recent advances in atomic nanofabrication with laser manipulated atomic beams are discussed with regard to the creation of arbitrary structures.

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1. Introduction

Continued miniaturization of electronic, optical and many other devices has a general and natural limit when the size of individual atoms is reached. Optical lithography as well as electron beam lithography will most likely advance technology to this limit between 2010 and 2020. When this limit is approached, the interesting question arises whether the number of atoms introduced into materials in order to control their functionality may have to be extended beyond the standard limit set by Poissonian statistics. For instance, stochastic composition of materials or doping with a small, random number of atoms may no longer be acceptable since it will cause large fluctuations of electronic or other properties once the scale of individual atoms is resolved. Advanced proposals in another field of research for the realization of solid state based quantum information processing also rely on the preparation of isolated atoms in a host material, demanding novel engineering concepts at the atomic scale.

Engineering at the atomic scale has been already realized nearly 2 decades now with scanning probe technologies, including the celebrated electron corrals and other beautiful structures. With these methods atomic structures are typically engineered on well prepared surfaces with atoms or molecules already present on this surface, while controlled transport of atoms or molecules towards the surface and in very small quantities is not directly part of these concepts. In atom lithography [1, 2], or more appropriately atom nanofabrication (ANF) [3], precisely this goal is achieved - the deposition of atoms transported to surfaces is controlled by means of laser light. In this report we will give a brief account of the current status of ANF and speculate about its future evolution at the ultimate atomic level.

2. Short Overview: Atomic Nanofabrication

In ANF atoms are transported to a surface through an atomic beam which is derived from a conventional thermal source, from a magneto-optical trap ('2D$^+$-MOT', [4]), or even formed as a coherent matter wave ('atomlaser', [5]) emanating from an atomic Bose-Einstein condensate. While the 2D$^+$-MOT as well as the atomlaser allow very tight control of atomic motion they suffer from relatively small flux densities. This is detrimental if large areas are considered for

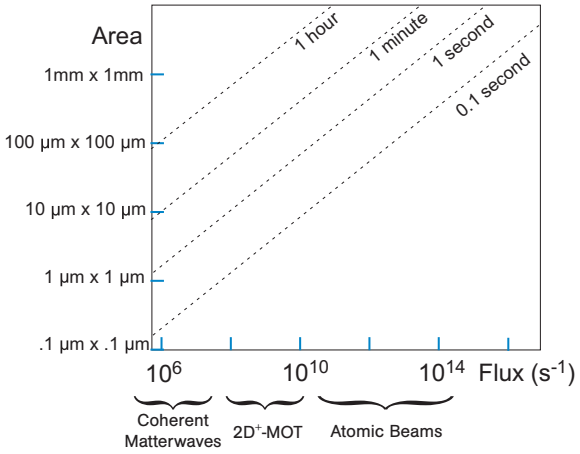

Figure 1. In atomic nanofabrication an atomic beam transports atom onto surfaces. The dashed lines in this graph indicate the time it takes to cover a given area with typical atomic beam sources.

parallel deposition but may not be a real obstacle when extremely small numbers of atoms have to be transported to a surface. In fact, a deterministic source of slow individual atoms has been realized with rates up to 10 kHz already [6, 7] as a first step towards controlled atom sources.

In every molecular beam epitaxy (MBE) apparatus a stream of atoms is transported to a substrate in order to grow a structure layer by layer and hence allow to control the vertical composition of the system. In ANF, also the uniform density of the atomic beam is modulated by transversal light forces derived from laser light interacting with the atoms. The resulting pattern is then transferred to the substrate. In contrast to MBE the growth of lateral structures in ANF requires local atom-surface interactions with low atom mobility at the surface. Since the light forces are derived from interfering light fields, the scale of the resulting patterns can be very naturally chosen at the sub-wavelength scale.

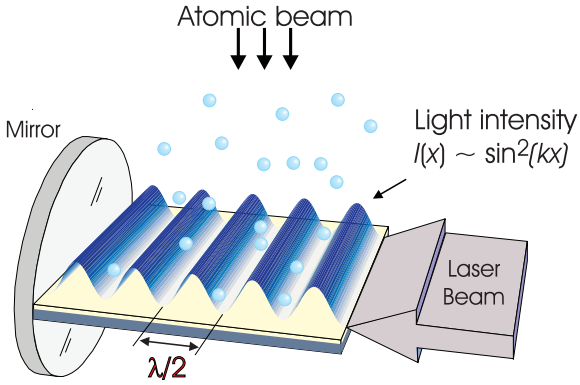

Figure 2. The well known concept of atom lithography: An atomic beam travelling through a standing wave light field towards a substrate is focused into narrow lines. If the laser is blue (red) detuned from the optical transition frequency optical dipole forces are exerted towards the nodes (antinodes) of the light field.

The origin of the action of a light mask onto the motion of atoms are optical dipole forces. In a laser beam with inhomogeneous intensity distribution $I(x)$ such as in a standing wave, an atom travelling in this light field experiences a force

$$\mathbf{F}(x) = -\frac{\hbar\gamma^2}{8\delta I_{sat}}\,\boldsymbol{\nabla} I(x) \tag{1}$$

where $I_{sat} = \pi hc/3\lambda^3\tau$ is the saturation intensity at the atomic resonance wavelength λ, $\tau \equiv 1/\gamma$ is the atomic excited state lifetime, and $\delta = \omega_{\mathrm{Laser}} - \omega_{\mathrm{Atom}}$ is the detuning.

We can distinguish two general methods of ANF: The most interesting one for applications grows lateral structures on a substrate by directly depositing atoms ('Direct Deposition', DD).

This method is very attractive since no other method is known which could laterally control growth procedures at the nanometer scale in a single step. Furthermore the method promises generation of 3D structures [15] when in addition to the lateral modulation of the atomic beam density the flux of the constituents of the atomic beam is regulated.

In the second method which is called 'Neutral Atom Lithography' (NAL) atom beams are used to modify the chemical properties of certain surfaces acting as a 'resist'. About a decade ago it was found that combinations of metastable rare gas [8]and alkali atomic beams [9] with thiole covered gold surfaces (SAMs, self-assembled monolayers) offer convenient and robust systems for this purpose. After exposure to such an atomic beam with modulated transverse density the structures can be prepared by wet chemical etching procedures. Both methods have shown to generate structural features below 100 nm and down to the 30 nm range [10, 11, 12].

For the successful application of ANF two main aspects have to be considered: (1) The role of laser cooling. (2) The atom-surface interaction.

2.1. The Role of Laser Cooling for ANF

The application of the feeble light forces [13] requires small atomic kinetic energies, thus demanding excellent collimation of the atomic beam. Excellent collimation can be obtained with conventional thermal sources by building long apparatus and using tiny apertures which obviously causes very small flux densities and hence long exposure times. Laser cooling overcomes this problem by reducing the divergence angle to below 1 mrad typically. In comparison with geometric collimation this method provides a flux enhancement exceeding two orders of magnitude easily. A drawback is of course that laser cooling must be realized before it can be applied. Most atoms susceptible for laser cooling to date such as the alkali and rare gas atoms are of more academic interest. Atoms of technological relevance and shown to be applicable include Cr, Yb, Al, Ag, and Fe, while research is underway to also laser cool Ga, In, and Er.

2.2. Atom-Surface Interaction in ANF

For ANF it is essential that the lateral structure encoded into the atomic beam survives impact onto the surface. For direct deposition an important requirement is thus negligible surface diffusion since otherwise structures were washed out. It is not known what the properties of the crystal growth under such conditions are to date.

For NAL lithography on the other hand, chemical reaction mechanisms (atom-resist) with high efficiency are required. It was found that typically 1-10 atoms are needed in order to cause a chemical modification making the thiole-resists susceptible for wet etching. A detailed understanding of the chemical mechanism ruling this interaction is still not available [14], but it is interesting to note that not only highly reactive species such as metastable rare gas atoms or alkali atoms but also Ba, Ga, and In show similar behaviour.

3. Parallel deposition of neutral atoms and complex patterns

One of the interesting aspects of ANF is the parallel generation of nanostructures across a relatively large area. It has been shown that a light mask with 1D and 2D periodic and non-periodic interference patterns can be imaged by the ANF methods. Since the generated pattern reflects the geometric intensity distribution of the interference field of the light mask, the degree of complexity which can be achieved depends on the complexity that can be written into the interference field. Although superposition of multiple light waves for this purpose has been experimentally demonstrated with a 'holographic mirror' [18], it is also known that the construction of a given intensity pattern from interfering field amplitude is a very difficult and in general unsolved mathematical problem [19]. In a recent proposal [20] we have suggested to overcome this problem by superposing standing light waves with frequency differences of the

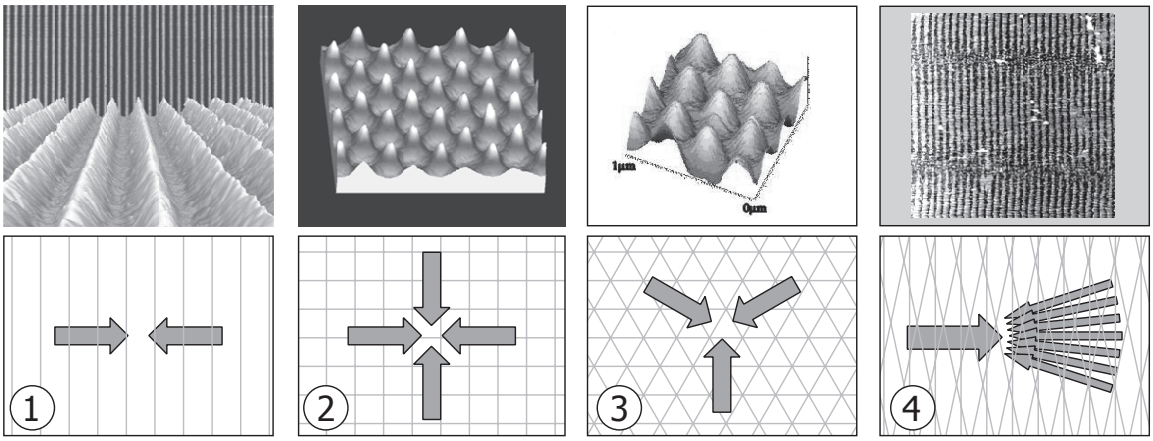

Figure 3. Gallery of 2D patterns generated by ANF. References: (1) [2]; (2) [16]; (3) [17]; (4) [18]

order of 100 MHz. Since atomic motion is insensitive to field variations at this frequency, it is only the superposition of intensities rather than field amplitudes which causes the atomic beam density modulation. In Fig. 3 a computer simulation is shown which assembles letters A,B,C,... from 24 laser beams which can be taken from a single source by frequency or time multiplexing. [19]

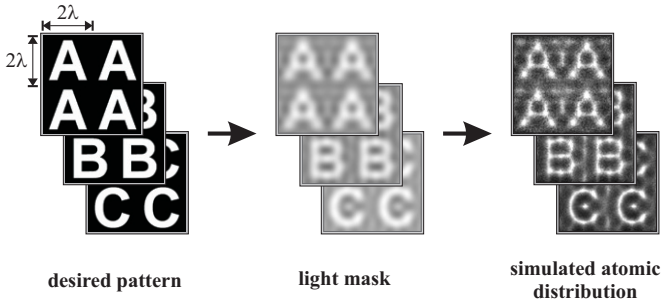

Figure 4. ANF generation of letters from a superposition of 24 standing wave light fields with different frequencies. Computer simulation from [20]

Currently research with parallel deposition concentrates on the generation of periodic structures with technologically relevant materials. For instance, laser cooling as well as writing of simple structures has now been demonstrated with iron atoms which is of interest for the creation of magnetic nanostructures [21, 22].

4. Serial deposition of neutral atoms - an atom pencil

Versatile deposition of atoms into any structural shape can straightforwardly be realized by constructing an atomic pencil where the flux of an atomic beam is directed into a single spot on a given substrate. While the speed of this method is of course much slower than the parallel method of the preceding paragraph, it maintains specific advantages of atomlithography such as direct deposition of atoms onto surfaces at low energy impact. The challenge of this flexible method lies in the creation of small spot or feature sizes while providing reasonably short exposure times, i. e. a reasonable flux density. A shadow mask approach has previously been used by Lüthi et al. [23] to create nanostructures by direct deposition from an effusive atomic beam source collimated by purely geometric means. In this experiment the source had to be mounted very close to the substrate in order to achieve significant flux density, a restriction which is overcome by the application of the transversely laser cooled beam.

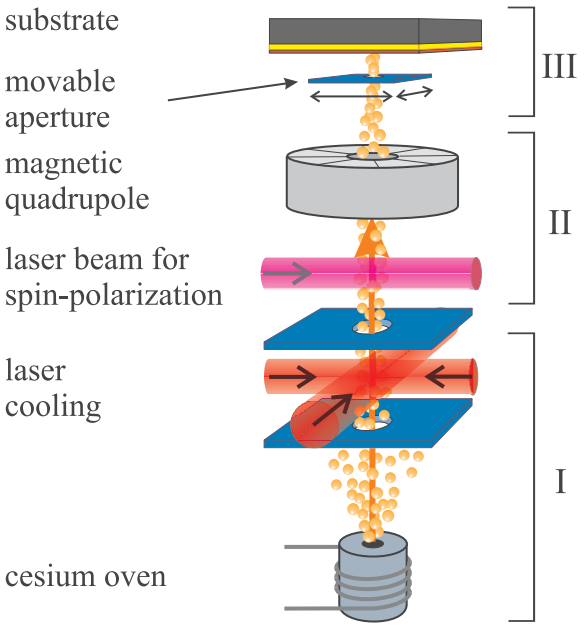

Figure 5. Setup for the atom pencil device. Laser cooling effectively enhances the flux density at the substrate by up to 2 orders of magnitude. A magnetic 4-pole (length and inner bore diameter 20 mm) further concentrates atomic flux onto a movable aperture which is used to create the desired structure, generating in our case another 40-fold increase of the flux density. In contrast to laser cooling this enhancement is achieved at the expense of increased divergence which sets stringent limitations on the separation of the aperture and the substrate.

Concentration of an atomic beam into a sub-wavelength area is a longstanding goal but difficult to achieve by purely optical forces in a focused light field only. We have thus chosen [24] a combination of a laser cooled atomic beam with a shadow mask as shown in Fig. 4.

In order to further enhance the flux density we have chosen to apply magnetic forces directing atomic trajectories - after optically pumping atoms into the most sensitive magnetic sublevel - towards the axis of the atomic beam and hence the aperture. A plane magnetic 4-pole ($|B_4(r)| \propto r$), which is conveniently constructed from permanent magnetic materials offering strong field gradients [25] is the equivalent of an optical axicon and a 6-pole ($|B_6(r)| \propto r^2$) is the equivalent of the optical lens.

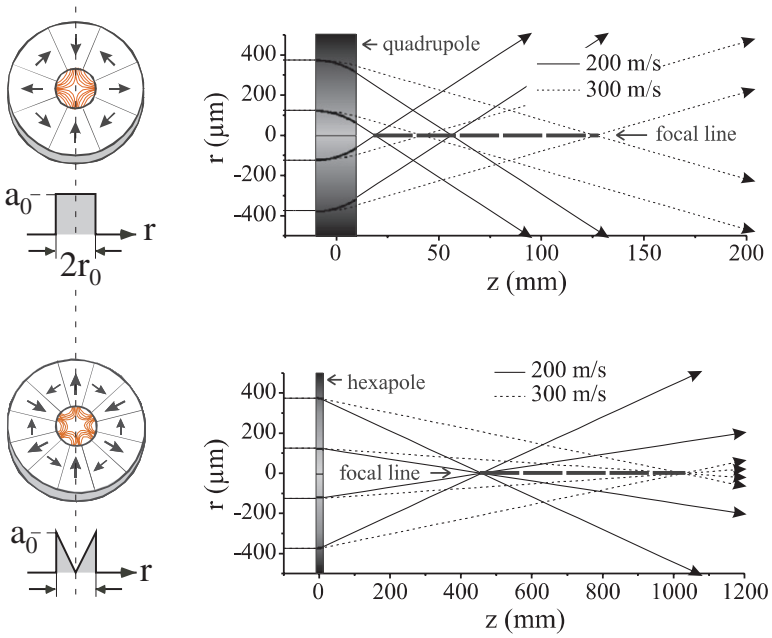

Figure 6. Magnetic 4-poles and 6-poles and their concentrating action on atomic trajectories. The 4-pole is the equivalent of an optical axicon, where parallel incoming trajectories of mono-velocity atoms are bent into a series of parallel outgoing trajectories. The 6-pole is equivalent to the optical lens and

While at first sight the magnetic 6-pole lens seems the obvious choice for optimum

concentration of the atomic beam it turns out that the axicon serves this purpose for a thermal atomic beam indeed much better. The reason is that the deflection depends on the longitudinal atomic velocity, and hence a focal line instead of a focal spot is produced not only by the axicon but also by hexapole. More detailed analysis shows [24] that the 4-pole has a characteristic 'focal' length $z_4 = Rv_{\rm th}^2/a_4L$ where R is the initial diameter of the incoming beam, L is the length of the 4-pole, $v_{\rm th}$ is the most probable thermal velocity, and $a_4 = \mu\partial_r|B_4(r)|/M$ is the 4-pole radial acceleration for an atom with magnetic moment μ and mass M. A similar analysis for the 6-pole constructed from the same material and for similar geometric conditions yields with $a_6 = \mu R\partial_r^2|B_6(r)|/M$ that $z_6 \approx 6 \cdot z_4$, or 130 and 740 mm in our experiment. The axial flux density can be calculated analytically yielding the dependence with normalized separation from the concentration multipole shown in Fig. 4. At short working distances from the concentrator the 4-pole is obviously superior to the 6-pole.

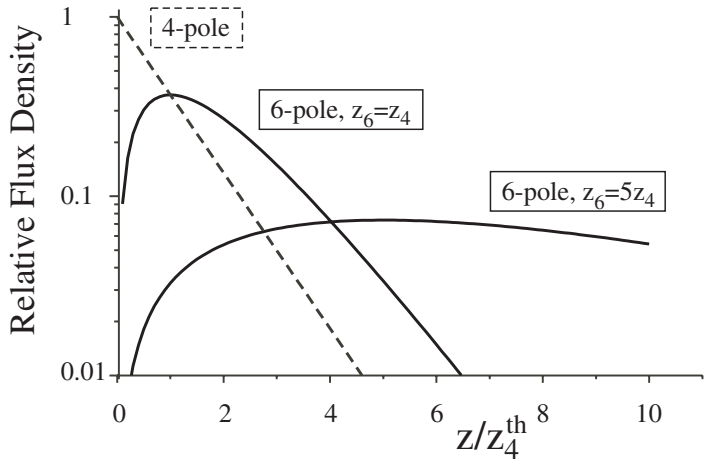

Figure 7. Comparison of axial flux density enhancement of a paramagnetic atomic beam by a magnetic 4-pole and a 6-pole with identical and with five times larger characteristic focal length.

We have applied the atom pencil with a Cesium atomic beam directed through a shadow mask as small as about 0.4 μm onto a SAM covered gold surface. The structure was in a final step prepared by wet etching procedures. In a separate experiment our colleagues at the University of Toulouse [26] have observed the clogging behaviour of the apertures which became noticable for the smallest apertures below 300 nm opening. They also showed, however, that heating of the aperture largely removes this problem. Furthermore this work contains interesting aspects about the use of SAMs as resist for neutral atom lithography.

5. Future Directions of Atom Deposition

Both the serial and the parallel approach to atom deposition presented here rely to date on atom sources subject to Poissonian statistics. At the current level where typically large numbers of atoms are deposited this does not present any problem. However there is growing interest in methods to deposit small and precisely controlled amounts of atoms into well defined areas. An interesting step into this direction was taken by the development of deterministic sources of individual atoms which have already been demonstrate to operate up to rates of 10^4 atoms per second. At very low rates collateral contamination of the substrate with non-controlled ambient atoms of the same kind will pose a serious problem. It may be of interest to develop tight atom guides such as offered by guides constructed with magnetic or optical forces in order to transport atoms with ultimate control - i. e. atom by atom - from a preparation device to a clean substrate for generating functional devices.

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References

[1] G. Timp, R. E. Behringer, D. M. Tennant, J. E. Cunningham, M. Prentiss, K. K. Berggren, Phys. Rev. Lett. **69**, 1636 (1992)
[2] J. J. McClelland, R. E. Scholten, E. C. Palm, R. J. Celotta, Science **262**, 877 (1993)
[3] D. Meschede, H. Metcalf, J. Phys. D **36**, R17-R36 (2003)
[4] K. Dieckmann, R.J.C. Spreeuw, M. Weidemller and J.T.M. Walraven Physical Review A **58** 3891 (1998)
[5] I. Bloch, M.Kohl, M. Greiner, T.W. Hänsch, and T. Esslinger Phys. Rev. Lett. **87** 030401 (2001)
[6] S. Kuhr, W. Alt, D. Schrader, M. Mller,V. Gomer, D. Meschede Science 293, p. 278-280, (2001),
[7] S.B. Hill and J.J. McClelland, Appl. Phys. Lett. **82**, 3028 (2003)
[8] K.K. Berggren,A. Bard, J.L. Wilbur, J.D. Gillaspy, A.G. Helg, J.J. McClelland S.L. Rolston, W.D. Phillips, M. Prentiss, and G.M. Whitesides Science **269** 1255 (1995)
[9] M. Kreis, F. Lison, D. Haubrich, D. Meschede, S. Nowak, T. Pfau, and J. Mlynek, Appl. Phys. B **63**, 649 (1996)
[10] W.R. Anderson, C.C. Bradley, J.J. McClelland, and R.J. Celotta Phys. Rev. A **59** 2476 (1999)
[11] F. Lison, D. Haubrich, P. Schuh, and D. Meschede Appl. Phys. B **65** 419(1997)
[12] A. Camposeo, F. Cervelli, F. Tantussi, M. Lindholdt, F. Fuso, M. Allegrini and E.Arimondo, Materials Science & Engineering C **23**, 1087 (2003)
[13] H. Metcalf, P. van der Straaten, Springer New York (1999)
[14] C. di Valentin, A. Scagnelli, and G. Pacchioni J.Phys. Chem. B **109** 1815 (2005)
[15] M. Oberthaler, T. Pfau J. Phys.: Condens. Matter **15**, R233 (2003)
[16] R. Gupta, J.J. McClelland, Z.J. Jabbour, and R. Celotta Appl. Phys. Lett. **67** 1378 (1995)
[17] U. Drodofsky, J. Stuhler, T. Schulze, M. Drewsen, B. Brezger, T. Pfau, and J. Mlynek Appl. Phys. B **65** 755 (1997)
[18] M. Mützel, S. Tandler, D. Haubrich, D. Meschede, K. Peithmann, M. Flaspöhler, and K. Buse, Phys. Rev. Lett. **88**, 083601 (2002)
[19] M. Mützel, U. Rasbach, D. Meschede, C. Burstedde, J. Braun, A. Kunoth, K. Peithmann, K. Buse, Appl. Phys. B **77**, 19
[20] M. Mützel, D. Meschede Laser Physics, **15**, 95 (2005)
[21] G. Myszkiewicz, J. Hohlfeld, A. J. Toonen, A. F. van Etteger, O. I. Shklyarevskii, W. L. Meerts, and Th. Rasing, Appl. Phys. Lett. **85**, 3482 (2004)
[22] 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, Appl. Phys. Lett. **85**, 4493 (2004)
[23] R. Lüthi, R. R. Schlittler, J. Brugger, P. Vettiger, M. E, Welland, J. K. Gimzewski, Appl. Phys. Lett. **75**, 1314 (1999)
[24] M. Mützel, M. Müller, D. Haubrich, U. Rasbach, and D. Meschede. C. O'Dwyer, G. Gay, B. Viaris de Lesegno, J. Weiner, K. Ludolph, G. Georgiev, and E. Oesterschulze, Appl. Phys. B **80**, 941 (2005)
[25] K. Halbach, Nucl. Inst. Meth. **169**, 1 (1980)
[26] C. ODwyer, G. Gay, B. Viaris de Lesegno, J. Weiner, K. Ludolph, G. Georgiev, E. Osterschulze, J. Appl. Phys. **97**, 114309 (2005)