High capacity tagging using nanostructured diffraction barcodes

G. S. Galitonov, S. W. Birtwell, and N. I. Zheludev

EPSRC Nanophotonics Portfolio Centre, School of Physics and Astronomy, University of Southampton, Southampton, SO17 1BJ, UK

swb@phys.soton.ac.uk

H. Morgan

School of Electronics and Computer Science, University of Southampton, Southampton, SO17 1BJ, UK

Abstract: We describe a new non-contact high capacity optical tagging technique based on the use of nanostructured barcodes. The tags are generated from a number of superimposed diffraction gratings. Capacity for up to 68,000 distinguishable tags has been demonstrated, however current technological capability shall allow encoding of up to 10^9 distinguishable particles, each of which is only 100μm long.

OCIS codes: (050.1970) Diffractive optics; (000.1570) Chemistry; (000.1430) Biology and medicine

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The rapid advances in high throughput screening, combinatorial chemistry, genomic and proteomic sciences have stimulated dramatic development of new encoding strategies for bead-based assays. Several optical encoding methods are currently used in these applications [1],

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including fluorescence, IR [2], Raman [3] and optical [4, 5] tagging on microbeads. Methods using magnetic tagging are also being investigated [6]. In particular, multicolor optical coding has been achieved by embedding quantum dots of zinc sulfide-capped cadmium selenide nanocrystals into beads [7-10] while patterns can also be written in fluorescently dyed beads by spatially selective photo-bleaching to create spatially selective fluorescent tags [11]. Here we describe a new method for encoding small beads which allows for non-contact reading. The tagging technique is based on fabricating a nano-structured pattern on the surface of the particle, which is only a few microns in size. The pattern is read by detecting the spatial distribution of laser light diffracted by the tag. Encoding information on the tag therefore requires creating many different patterns ,which can produce large numbers of unique distributions of diffracted light. In the simplest implementation, the pattern is no more than a miniature diffraction grating, where information is coded in the pitch or spatial dimensions of the grating. However, the encoding capacity of such a tag can be greatly increased by fabricating tags with several overlapping nano-scale gratings.

In a classical single diffraction grating, information may be encoded in the pitch or spacing of the grating, a. When such a grating is illuminated with light at wavelength λ, for example at normal incidence, a series of diffracted beams of different order m is created, according to the equation $a\sin\alpha = m\lambda$. Here α is the angle of diffraction and m is the diffraction order. Therefore, a measurement of the first order diffraction angle, at $m = 1$, gives direct information about the pitch of the grating a. This principle is shown in Fig. 1. The number of distinguishable tags that can be manufactured depends on the ability to resolve a difference between two diffraction patterns. Two diffracted beams at diffraction angles θ_1 and θ_2 can be resolved if $|\theta_1 - \theta_2| \geq \Delta\theta_1/2 + \Delta\theta_2/2$, where $\Delta\theta$ is angular width of the beam. Here $\Delta\theta = \lambda(Na\cos\theta)^{-1}$, where N is the number of periods in the grating of length L ($N \approx L/a$). Using this criterion it is possible to calculate the capacity of diffractive bar-codes with a single grating as a function of the grating length (neglecting all diffraction orders above $m = 1$). The results are presented in Fig. 2, curve (1).

A much larger capacity of tag can be obtained if several gratings are superimposed on the bead, as shown on Fig. 2. Each of the superimposed gratings produces its own set of diffracted beams independently of the other gratings on the tag. If the tag has k superimposed gratings, the number of possible distinguishable codes c is then given by the number of possible combinations of k beams in n possible positions, using the combinatorial formula $c = \frac{n!}{(n-k)!k!}$. The results of calculations of the tag capacity for several superimposed gratings using the above resolution criteria is presented by curves (2)-(5), Fig. 2. In practice the maximum number k_{max} of superimposed gratings in the tag which can be distinguished by diffraction is limited by the resolution δ of the tag's fabrication process and may be estimated as $k_{max} \simeq \lambda/(2\delta)$.

This analysis assumes that the data is encoded only in the first order diffraction spots. In practice, however, higher diffraction orders will be present, which could be confused with the first order beams, leading to misreading of the tag. In general intensities of high-order diffraction beams depend on the grating aspect ratio and in practice are much smaller than intensities of the first order beams. For instance a grating in which transparent and non-transparent strip are of the same width does not produce any second order diffraction beams. However, for the purpose of identification it is critical to be able to distinguish the first order diffraction beams from the higher order beams. This can be achieved by use of intensity discrimination of the higher order diffracted beams, which are normally less intense than the first order diffraction. The required intensity discrimination threshold depends on the physical characteristics of the diffraction grating, in particular on the ratio $a/(a-b)$, where b is width of the transparent grating elements (see Fig. 1). The discrimination threshold level S was calculated for an ideal grating made of perfectly transparent and perfectly opaque strips for different values of $a/(a-b)$, by calculating

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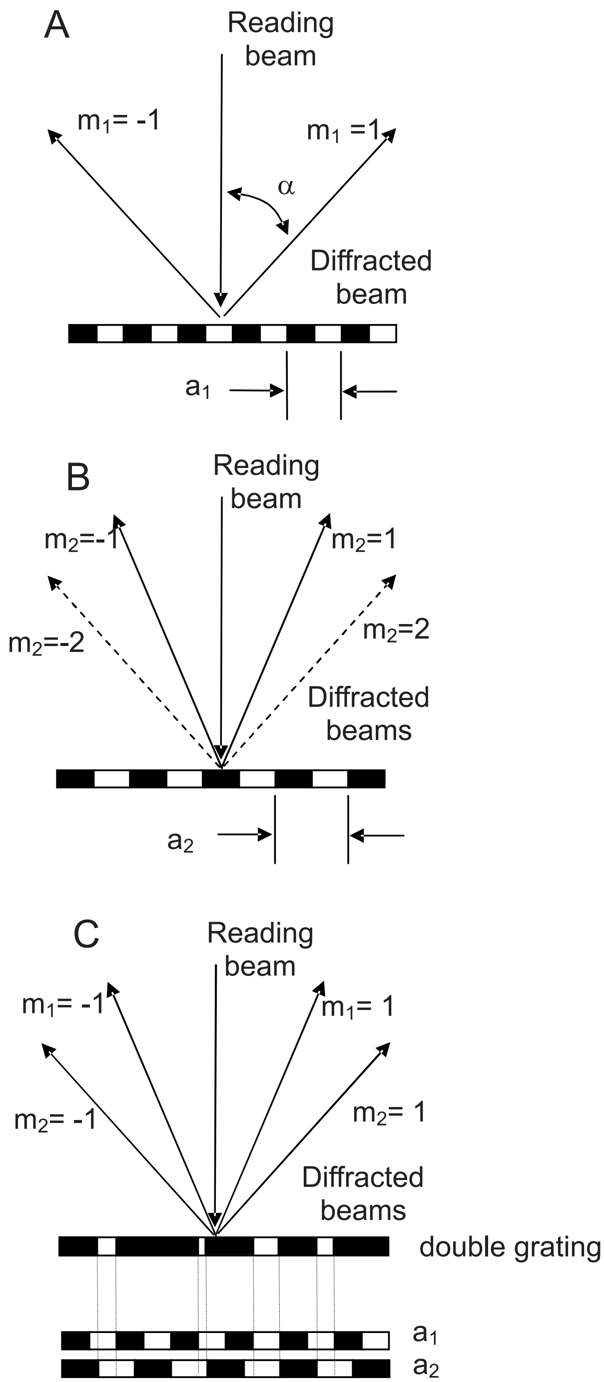

Fig. 1. A) first order diffraction from grating with pitch a_1; B) first and second order diffraction from grating with pitch a_2. Higher order diffraction is normally less intense and for encoding applications higher orders may be eliminated by threshold detection; C) first order diffraction from combinatorial grating made by superposition of gratings with pitches a_1 and a_2

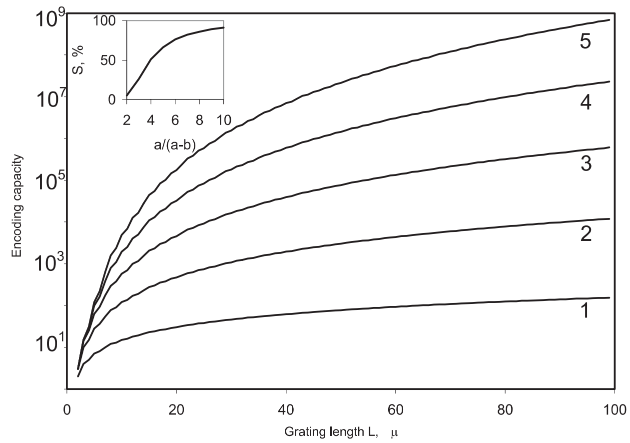

Fig. 2. Encoding capacity of a diffractive bar-code tag as a function of the length of the tag for different numbers of superimposed gratings. The number on each curve corresponds to the number of superimposed gratings. The inset shows the level of intensity discrimination *S* necessary to ensure error-free identification of tags

the intensity of the brightest higher order diffracted beam as a fraction of the first order beam intensity. This shows that gratings with $b = a/2$ require the lowest level of threshold, and setting S = 0.05 eliminates all higher diffraction order beams and provides error-free identification of the tag (see inset of Fig. 2).

Some further reading problems can appear when gratings are superimposed: although far-field Fresnel diffraction as a Fourier transform of the field distribution on the grating is a linear process, superposition of gratings is not exactly equivalent to creating a linear sum of their fields. This could lead to interplay between gratings and appearance of extra ’ghost’ beams not present in individual gratings. Our modeling shows that intensities of the host beams are generally much smaller than that of the main beams. They depend on the aspect ratio of the gratings and could become a serious factor at high levels of superposition. Tag capacity limitations arising from the ghost beams for high order tags will have to be investigated further, but does not appear to be a major factor in the experiments reported below.

In order to demonstrate this tagging concept a chip library of chromium gratings was manufactured on a glass substrate using direct write electron beam lithography. The library of gratings contains almost 7,400 unique barcode tags, 50x50 μm, separated by gaps of 200 μm. SEM images of these tags showing the range of different superimposed gratings are presented in Fig. 3. With an available nanofabrication resolution δ of about 100nm we have been able to demonstrate tags up to order three (containing three superimposed gratings) that are fully distinguishable by diffraction. This provides a capacity of about 68,000 distinguishable tags. Higher order tags have also been fabricated, but they sometimes show fails in pattern reproduction which spoils the quality of diffraction (Fig. 3(iv))

An example of the diffraction patterns created by these tags is presented in Fig. 4. The gratings were read using light from a HeNe laser (633nm) incident at normal angle to the sample. The diffraction pattern was observed on a screen parallel to the grating and captured using a CCD array detector. Fig. 4(a) shows how the diffraction pattern changes with grating pitch; increasing in complexity from the simplest single grating tag. The series of diffraction patterns (A to J) demonstrates how it is possible to uniquely distinguish between ten different tags containing only a single grating. In the photographs, the first order diffraction spots are highlighted by the solid circles, while the positions of the much weaker second order diffraction spots is

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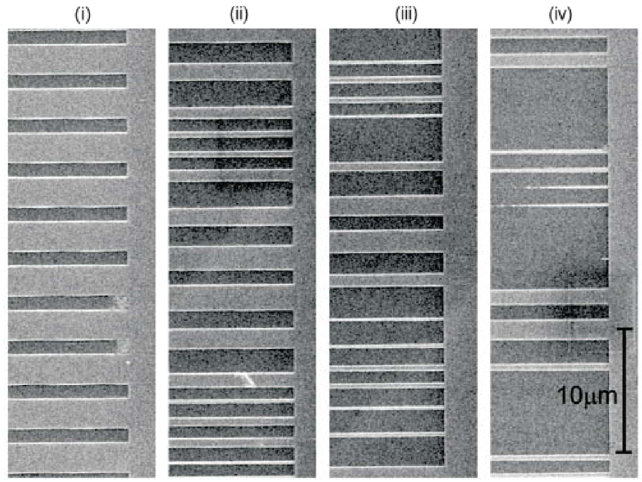

Fig. 3. SEM images of barcode tags of different order: (i) single grating tag; (ii) two superimposed gratings; (iii) three superimposed gratings; (iv) four superimposed gratings (note nano-lithography resolution limiting quality of this grating).

indicated by dashed circles.

Figure 4(b) shows diffraction patterns from different tags containing three superimposed gratings (patterns HFG to QFG). Here, the first grating diffracts exactly as grating G in Fig. 4(a), while the second grating diffracts as grating F. The third grating differs from pattern to pattern. The first diffraction pattern is from a grating which diffracts as grating H in Fig. 4(a), but then changes step by step to give a higher and higher diffraction angle. In this way we have resolved diffraction patterns with up to three superimposed gratings. Further increase in the number of superimposed gratings on the tags led to increasing read errors due to the limited resolution of the grating fabrication process.

In conclusion we have demonstrated a new optical, non-contact tagging technique based on superimposing large numbers of miniature diffraction gratings on a tag. With a 50nm nanofabrication resolution now routinely available, the technique is capable of creating distinguishable tags containing at least 5 superimposed grating and encoding up to 10^9 tags, each of which is only 100μm long and a few μm wide. To demonstrate this technique we manufactured a library of 50μm x50μm tags on a glass wafer. We have been able to demonstrate experimentally that the principle of superimposing works. With nanofabrication resolution of about 100nm it has been possible to resolve tags containing at least three superimposed gratings providing capacity for more than 68,000 tags. An enormous increase in capacity will be possible if two sets of mutually perpendicular gratings are used. Combinatorial analysis shows that up to 10^{18} different barcode tags can possibly be fabricated with such two-dimensional superimposed gratings up to order five. Although the principle of the technique has been demonstrated by fabricating metal-on-glass tags, equally the tags could be fabricated onto a polymer material such as a small micron sized particle. The robust nature of the tags, together with the non-contact remote reading capability makes them ideal for a large variety of biochemical, cytological, proteomic and genomic applications. These tags could also have widespread use in invisible marking for security applications and product marking identification and tracking.

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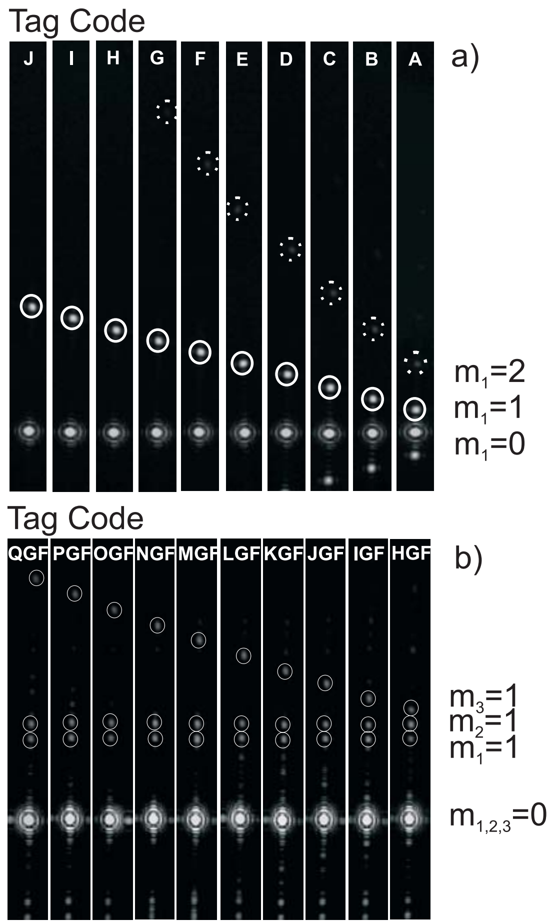

Fig. 4. Diffraction patterns created by a single grating tag (a), and tags containing three different gratings (b). Moving from left to right shows how a progressive decrease in the pitch of one of the gratings changes the diffraction pattern

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