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Optical diffraction by ordered 2D arrays of silica microspheres

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ABSTRACT

The article presents experimental and theoretical studies of angular dependent diffraction properties of 2D monolayer arrays of silica microspheres. High-quality large area defect-free monolayers of 1 μm diameter silica microspheres were deposited by the Langmuir-Blodgett technique under an accurate optical control. Measured angular dependencies of zeroth and one of the first order diffraction efficiencies produced by deposited samples were simulated by the rigorous Generalized Source Method taking into account particle size dispersion and lattice nonideality.

1. Introduction

Ordered monolayer arrays of silica microspheres attract high interest due to promising applications in photovoltaics, mainly for light trapping functionality [1]. They were demonstrated to increase outcoupling efficiency in organic light-emitting diodes [2] and to possess attractable antireflection properties useful for solar cells [3]. Layers of silica microspheres often represent highly ordered structures thus making diffracted light to be strongly angular dependent. For applications it is then important to be able to carefully predict and optimize both spectral and angular optical characteristics of arranged silica particles. From manufacturing viewpoint, quality of microsphere structures and density of defects they contain dramatically depend on a fabrication process. Recently a diffraction pattern monitoring was proposed as means for in-situ optical control of fabrication process and structure quality [4–6]. Within such fabrication framework it is also interesting to study diffraction properties of silica microspheres arrays aiming at further improvement of such optical control. In addition to this in-situ diffraction based quality monitoring we are motivated by improvement of state-of-the art rigorous diffraction simulation for microsphere composed crossed gratings.

Experimental measurements of diffraction properties of ordered monolayers of dielectric microspheres in optical, terahertz, or microwave bands were reported by different researches. Authors of [6–8] studied far field directional properties of diffraction patterns appearing upon illumination of monolayers by laser radiation. Near field images can be found in [9]. Majority of researches studied spectral transmission and reflection of zeroth diffraction order [8–12]. In addition, angular resolved zeroth order diffraction efficiency can be found in [8,13]. Article [14] provides angular properties not only for the zeroth diffraction order, but also for first orders for particles with diameter-to-wavelength ratio $D/\lambda \approx 3$, though the experimental points are picked up with a rather large step. Authors also gave intuitive explanations for shapes of obtained angular dependencies. Article [15] demonstrates comparison between spectral reflectance properties of ordered and disordered monolayers.

Accurate simulation of experimental results on spectral and angular resolved transmission and reflection of various diffraction orders for microspheres with $D/\lambda \sim 1$ should be performed by a rigorous method. Several different approaches can be applied for this purpose. Various formulations of the modified Korringa-Kohn-Rostoker method were proposed and applied in [16–20]. Another surface integral based approach is given in [21] for diffraction calculation by a plane grating of dielectric bodies of revolution. Completely different class of low numerical complexity rigorous methods is based on two-dimensional Fourier representation of monolayers [22–24]. Our implementation of such Fourier approach is used in this work. Approximations for coherent reflection and transmission coefficients for a monolayer of spheres accounting for multiple reflections can be also obtained within the quasi-crystalline approximation [25,26]. This approximation was used for investigation of transmission and reflection spectra for imperfect lattices of spherical particles [27,28]. Also it is possible to

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perform rigorous diffraction simulations with the finite difference time domain, or the finite element methods [29–32].

In this work we supplement the mentioned experiments with carefully measured angular dependencies of diffraction efficiencies for zeroth and one of the first diffraction orders for 1 μm diameter microspheres illuminated with He-Ne laser beam, $\lambda = 0.6328$ μm. To rationalize the obtained data and trace changes in diffraction behaviour when passing from ideal to realistic monolayers we utilize rigorous diffraction simulation. For this purpose we applied the Generalized Source Method (GSM) [23] as it allows one to rigorously simulate diffraction on large period gratings with complex corrugation interfaces, to carefully control the accuracy of calculations, and it is extremely computationally efficient. Below we first describe sample preparation. Then we provide details on the experimental setup and a brief description of the GSM. And finally discussion of the results and conclusions are given.

2. Sample preparation

For preparation of experimental samples we used the Langmuir-Blodgett technique (Fig. 1). It is one of the most efficient methods for preparing regular monolayers of microspheres over large areas with homogeneous deposition of the particles. First, 1 μm diameter spherical silica particles were suspended in butanol (concentration was 35 mg/ml). After ultrasonic dispersion during 30 min, the particles were injected into a carrier fluid (moving water in Fig. 1) and brought to liquid surface to form a monolayer array with the aid of surface tension forces. Such monolayer is called “transfer zone”. The quality of the self-assembly of particles within the transfer zone was controlled by a camera with a custom image processing software [5,33]. The software monitors quality of the diffraction pattern, and distinguishes diffraction spots from blurred diffracted light. When an optimal layer quality is detected, BK7 substrates (5×7 cm^2, 3 mm thick) are lifted out of the fluid (at a speed of a few cm/min) and a monolayer of microspheres was transferred onto the substrates. Fig. 2 shows a substrate with an obtained film, and a typical electron microscope image of such film. Defect-free areas appear to have dimensions of dozens of micrometers, and the monolayer covers all substrate area.

3. Diffraction measurements

Experimental measurements of the diffraction efficiencies were taken with a setup demonstrated in Fig. 3. Linearly polarized collimated He-Ne laser beam of $\lambda = 0.6328$ μm wavelength diffracted on a substrate covered by a monolayer of 1 μm diameter silica particles described above. The substrate was fixed on a rotating platform, which allowed us to rotate the sample around vertical axis Y with accurate rotation control within 1°. The beam size was reduced in order to analyze defect-free areas of the monolayer, so that the latter can be considered to be close to a regular hexagonal

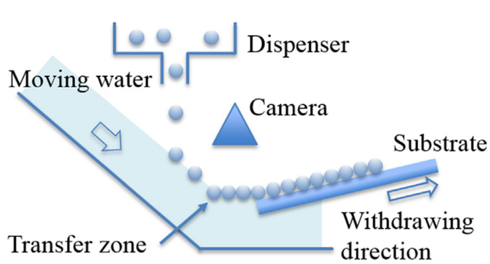

Fig. 1. Sketch of the Langmuir-Blodgett methods used for preparation of experimental sample of regular monolayer of silica microspheres.

arrangement of silica particles. In the experiment the beam spot diameter in the grating plane was several dozens of micrometers. Diffraction efficiencies were measured with a photodetector, which was placed several centimetres behind the substrate (image plane in Fig. 3) so as to capture the power of a corresponding diffraction order. Field of view of the photodetector was comparable to a size of diffraction spots and allowed us to analyze efficiency of each order separately. Fig. 3 also shows an image of the observed diffraction pattern. In this work we measured angular dependencies of nonvanishing $(0, 0)$ and $(-1, 0)$ diffraction orders as demonstrated in the figure.

4. Simulation

As was stated in the Introduction we used the GSM as currently it is one of the most efficient relevant numerical methods. The GSM allows one to rigorously calculate light diffraction on complex shape 2D periodic plane dielectric gratings of periods from tenths to dozens of wavelength. The method is based on the volume integral equation

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_{inc}(\mathbf{r}) + \omega^2\mu_0 \int \mathbf{G}(\mathbf{r} - \mathbf{r}')\left[\varepsilon(\mathbf{r}') - \varepsilon_b\right]\mathbf{E}(\mathbf{r}')d\mathbf{r}', \tag{1}$$

which is discretized in the 2D Fourier space reciprocal to the grating plane – plane XY in Fig. 3, and 1D coordinate space being perpendicular to the grating layer – Cartesian direction Z. This discretization of the integral along axis Z is conventionally called “slicing” of grating layer. Here $\mathbf{E}_{inc}(\mathbf{r})$ is the field generated by some external sources (e.g., a plane wave coming from infinity), $\mathbf{G}(\mathbf{r} - \mathbf{r}')$ is the Green tensor of free space, which should be written in the plane wave functional basis, $\varepsilon(\mathbf{r}')$ is spatially inhomogeneous permittivity in the grating region, and ε_b is a “basis” permittivity being a parameter of the GSM. Upon discretization Eq. (1) reduces to a linear algebraic equation system on unknown vector of diffraction order amplitudes. Number of equations in this system is proportional to product $N = N_{FX}N_{FY}N_S$, where $N_{FX}N_{FY}$ is the number of Fourier harmonics in the 2D Fourier space, and N_S is the number of slices used in discretization of the integral along coordinate space direction Z. Due to special properties of the equation system matrix this system can be solved with linear complexity relative to N by means of an iterative solver like the Bi-Conjugate Gradient or the Generalized Minimal Residual method. High parallelizability of the Fast Fourier Transform used in the system matrix by vector multiplications allows performing highly efficient computations on Graphical Processing Units (GPU). In addition, the S-vector algorithm [34,35] is used to accelerate convergence of a linear solver.

Presence of the substrate interface, which supports the monolayer, is rigorously handled within the GSM (see the sketch in Fig. 4(a)). Light interaction with the backside of the substrate was also taken into account using the technique described in [36] via power reflection and transmission coefficients for each plane wave. This simplification is possible since beam divergence and substrate thickness variations do not allow one to observe substrate interference effects. Additionally, for waves, which undergo multiple reflections inside the substrate, we treated the monolayer of microspheres as a homogeneous layer with volume average permittivity equal to 1.3^2 (i.e., waves reflected from the substrate backside and incidenting the monolayer from the substrate side do not diffract). This simplification dramatically reduces the required computation resource consumption, while having no visible impact on resulting angular dependencies of diffraction efficiencies. Within this approach substrate thickness and beam spot size were implicitly taken into account when estimating a number of reflections, which each plane wave undergoes in the substrate.