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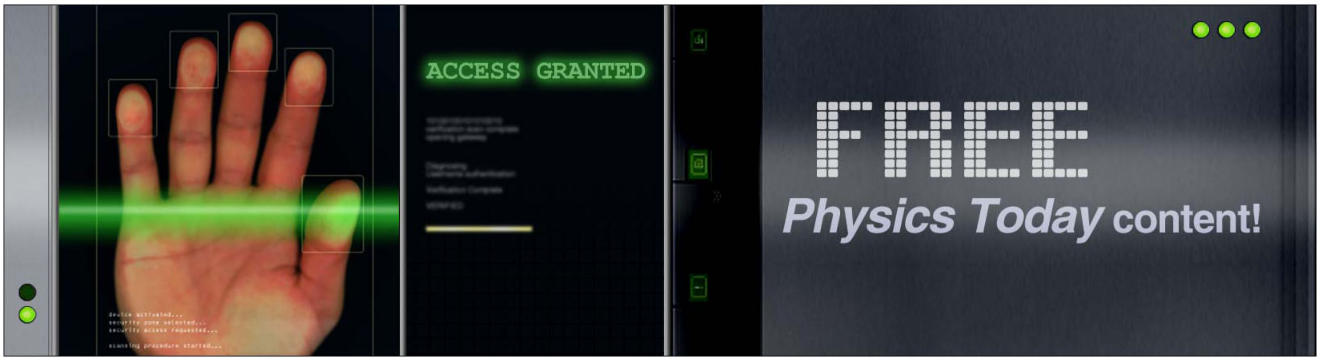

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Charge sensor and particle trap based on z-cut lithium niobate

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The generation of adhesive regions on a z-cut lithium niobate crystal without an additional voltage supply is demonstrated. We show that the origin of the attractive force in the respective solvent is electrophoresis, which can selectively trap charged particles in illuminated regions. Using digital holographic microscopy to measure the space-charge field in a y-cut crystal, we demonstrate the difference between electrophoretic and dielectrophoretic particle manipulation. The suggested method enables the creation of arbitrary two-dimensional patterns, circumventing restrictions originating from the crystal asymmetry. Furthermore, it allows the discrimination between charged particles of different signs, thus acting as a charge sensor. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4818143]

In the family of micromanipulation techniques, the field of high-throughput optical manipulation has long been the poor cousin of its much more famous relative, the optical tweezers technique. The holographic version of the optical gradient trap, as optical tweezers were first called,[1] is today able to confine a small number of particles in three dimensions and flexibly manipulate them independently.[2] However, there are some applications that request the ability to handle a very large number of particles distributed over a relatively large area, e.g., up to a few hundred by hundred micrometers.[3] Such requirements can hardly be simultaneously fulfilled by optical tweezers due to the high NA objectives necessary for trapping. Since the invention of opto-electronic tweezers (OETs), the field has seen quite a boost, allowing to use dielectrophoresis (DEP) to manipulate more particles at severely reduced light powers.[4] In particular, lithium niobate ($LiNbO_3$) has been suggested as a substrate material for OET, since it is able to generate the high electric field gradients necessary for efficient trapping by its outstanding photovoltaic capabilities.[5–8] Yet, a drawback of lithium niobate remains the anisotropy of the photovoltaic effect, which is at least one order of magnitude stronger along the c-axis of the material with respect to the perpendicular directions.[9] This results in a negligible trapping efficiency for grating vectors perpendicular to the distinguished axis,[10] effectively limiting the possible pattern configuration on lithium niobate substrates. In our contribution, we demonstrate that the use of a z-cut crystal can facilitate the creation of arbitrary trapping patterns on a $LiNbO_3$ surface without any restrictions due to crystal anisotropy. Most of the previous experiments on the applicability of lithium niobate as OET have used y-cut crystals to demonstrate and optimize optically induced trapping[5,10,11] or polymer shaping.[7] Instead, z-cut $LiNbO_3$ crystals (i.e., with the c-axis normal to the optical surface) have been employed for selective pyroelectric trapping at the edges of periodically poled $LiNbO_3$ samples.[12] The confinement in this case originates from dielectrophoretic forces created at the domain edges. Since in periodically poled $LiNbO_3$, the domains possess alternating directions of the crystal axis, high gradients of the electric field occur upon heating or cooling due to the crystal's pyroelectric properties. The origin of these forces is very similar to the ones in previous experiments with y-cut crystals, however, the trapping cannot be induced all-optically. Since the confinement of particles is limited to domain edges, the z-cut crystal must be appropriately poled beforehand to achieve a certain geometry, e.g., a hexagonal structure.[12] Although the sophisticated procedure of periodic poling of lithium niobate has matured into a standard technique in pure $LiNbO_3$ crystals,[13,14] it still remains difficult in doped samples.

In order to be able to take advantage of the beneficial features of lithium niobate samples, i.e., the absence of an external voltage supply and the all-optical configuration, yet to circumvent the problem of anisotropy, we suggest the exploitation of z-cut samples instead of y-cut ones. When a crystal in this geometry is illuminated by a laser beam, a charge separation is facilitated by the bulk photovoltaic effect. Since in $LiNbO_3$, the respective components of the photovoltaic tensor along most directions can be neglected, the main direction of charge transfer is along the c-axis of the material, which leads to the agglomeration of charges in the illuminated regions on the main faces of the crystal. Depending on the crystal orientation, these faces are denoted as +c or −c face. Among the mechanisms to manipulate matter, in this scenario, dielectrophoretic forces due to the electric field gradient between the illuminated and non-illuminated regions occur as the method of choice (see also Fig. 1). In previous experiments, however, it has been demonstrated that an electric field gradient sufficient for dielectrophoresis can only be induced along or in small angles to the c-axis[5,10] but not perpendicular to it, which means that in the z-cut geometry DEP is not to be expected. However, provided that particles in a suspension are charged, they can clearly be manipulated using electrophoretic interaction due to the occurrence of charged regions on the main faces of the crystal.

To prepare an adequate solution of particles, we use Novec 7500 (hydrofluorether, 3M) as a suspension medium. It was found empirically that when microparticles were suspended in this liquid, charges were acquired by these particles. In our experiments, we could see the effect for particle concentrations ranging from 1.5–12 $\mathrm{g\,l^{-1}}$. Due to the

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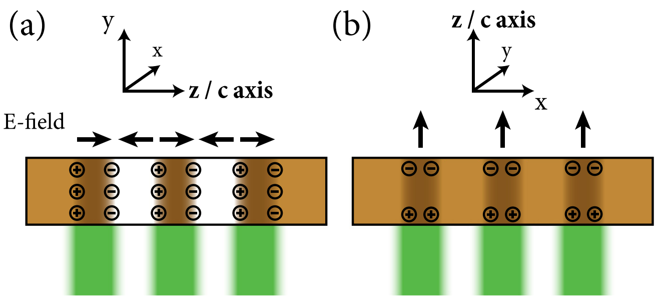

FIG. 1. Schematic for the difference between trapping on (a) y-cut and (b) z-cut crystals; in the case of a y-cut crystal, strong positive (dark) and negative (white) alternating electric fields along the surface provide high field gradients for dielectrophoretic trapping; in a z-cut geometry, a field normal to the surface is induced only for the illuminated regions, the resulting electric field gradients are substantially smaller.

fact that special care was taken not to insert charges from the outside, the most likely explanation for this behaviour is the transfer of charges from the polar solvent molecules through friction. In order to demonstrate the preparation of a solution of charged particles, we first use an Fe-doped y-cut crystal for trapping. In this geometry, the main field gradients are along the c-axis of the material, with the drawback of a strong material anisotropy. However, we can have both electrophoresis due to the occurrence of surface charges and dielectrophoresis due to the strong field gradients. The different particle trapping positions hence allow to discriminate between the two phenomena.

The crystal is integrated into a commercial microscope, where the rear port is used to couple in light of wavelength $\lambda = 532\,\mathrm{nm}$ into the body, modulated by a Full-HD resolution amplitude spatial light modulator (ASLM, *HED 6001* Holoeye) to obtain any desired light pattern. In order to correlate the electric field inside the material with the trapped particles' positions, the microscope is equipped with a Mach-Zehnder type interferometer to allow a digital holographic microscopy (DHM) measurement of phase retardations inside the material, as already applied previously in a different dielectrophoretic experiment.[6] A sinusoidal grating with a period of $\Lambda = 420\,\mu\mathrm{m}$ is induced inside the crystal (Fig. 2). The internal phase retardation and hence the electric field is acquired using DHM before particles in suspension are dripped on the crystal. For details about the assay, we refer to the previous literature.[6] The same trapping experiment is performed once with a suspension of glassy carbon (GC) powder (Ø2–12 μm, Sigma-Aldrich) in tetradecane, an unpolar, non-dissociating oil. Here, dielectrophoretic trapping is known to occur.[10] The second control experiment is with Novec 7500 as the suspension medium for the same particles and concentration of approximately $3\,\mathrm{mg\,ml}^{-1}$. Fig. 2 shows the internal space-charge field overlayed with the averaged particle density. The particle density in both cases is obtained from a simple white light image of the crystal surface, whose inverted grayscale is proportional to the particle density. Although particles lying on top of each other cannot be adequately resolved with this method, the position of particle maxima can still be reliably determined. Both values are acquired initially as two-dimensional images, however, for sake of better visibility, they have been averaged along the x-axis before plotting. In both cases, one can observe two lines of trapped particles within one sinusoidal electric grating period, but a fundamental difference lies in the position of the lines relative to the extrema of the electric field: In the case of tetradecane as the suspension medium, as in Fig. 2(b), particles come to rest at or in the vicinity of the electric field maxima and minima, where the gradient of the electric field is zero. Since the DEP force is proportional to the gradient of the electric field intensity, this complies very well with the theory of positive dielectrophoresis, which describes the particles as being attracted towards the regions of highest field intensity.[6,15] On the other hand, in the case of Novec 7500 as the surrounding medium (Fig. 2(c)), the particles align primarily at the positions of zero crossing of the internal space-charge field. From Gauss's law $\varepsilon_0 \nabla \vec{E} = \varrho$, it can be concluded that these are exactly the positions where the positive and negative charges responsible for the internal field modulation are located. Hence, we can deduce from this simple experiment that we have dielectrophoretic trapping in the case of tetradecane, whereas electrophoretic attraction dominates when Novec 7500 is used.

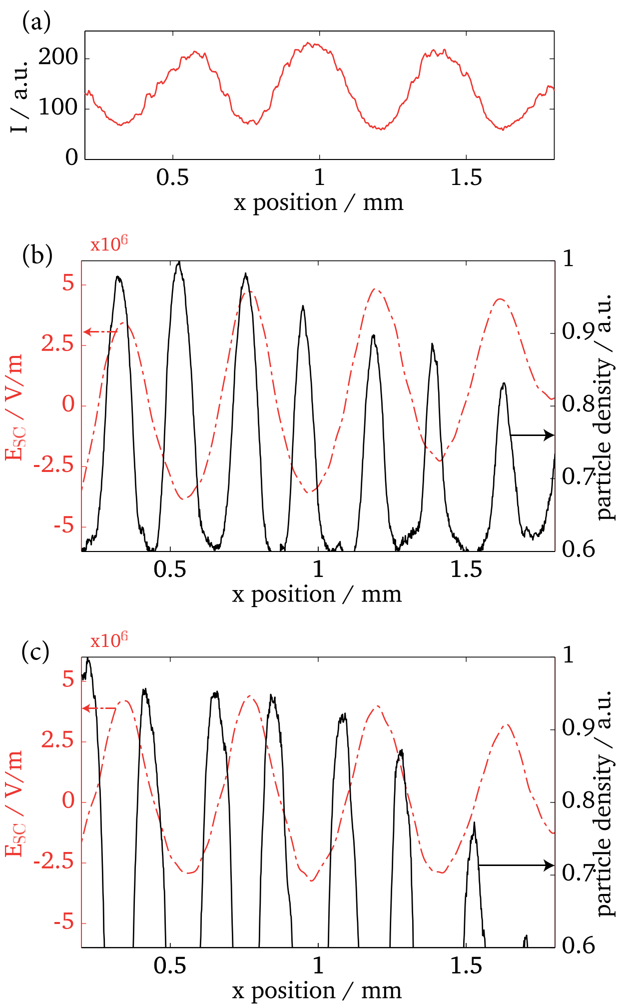

FIG. 2. Particle positions with respect to the internal space-charge field on a y-cut crystal: upon illumination with a sinusoidal intensity pattern (a), a space-charge field with π phase shift is formed inside the crystal (dashed red line in (b) and (c)); from the particle density (solid black line), it can be concluded that in the case of tetradecane as suspension medium (b), dielectrophoretic trapping dominates whereas electrophoretic trapping occurs for matter suspended in Novec 7500 (c); for details about the measurement refer to text.

After having shown that friction and collisions in engineered fluids can be used to reliably prepare a solution of charged particles, we can use such a solution to demonstrate optically induced trapping in arbitrary patterns in the z-cut geometry. To this end, the y-cut $LiNbO_3$ crystal is replaced by a z-cut sample with the same total iron dopant concentration of $18.8 \times 10^{18}\,\mathrm{cm}^{-3}$ and a reduction degree of $Fe^{2+}/Fe^{3+} = 0.16 \pm 0.01$.[8] As mentioned before, the illumination of the sample leads to a charge separation along the c-axis due to the bulk photovoltaic effect. Since it is not short-circuited, the agglomeration of charges on the main faces of the crystal can be used to attract microparticles of opposite charge. Fig. 3 shows the arrangement of GC particles suspended in Novec 7500 on the surface of a 900 μm thick z-cut crystal. The illumination pattern, sketched in the inset, contains all possible orientations of the light modulation and furthermore honors the 100th anniversary of Niels Bohr's discovery of the semi-classical shell model.[16] Prior to the addition of the suspension, the crystal has been illuminated with a low mean intensity of $8.9\,\mathrm{mW\,cm}^{-2}$ for 10 s, thereby inducing a charge pattern replicating the sketch. It is clearly visible that particles are attracted towards the illuminated regions and that, as expected, there is no anisotropy visible due to the rotational symmetry of the geometry. The sign of the charges that accumulate on the surface depends on the direction of the c-axis of the crystal, where the +c face of the crystal is charged negatively and the −c face charged positively.[16,17] In the case of Novec 7500, it can be concluded from Fig. 2 that both negatively and positively charged particles exist in the solution. This implies that even if the crystal is turned by 180°, the trapping results stay the same. It could be verified that the same alignment occurs with both orientations of the crystal c-axis. Once they are trapped on the crystal surface, the micro particles are reliably kept in place since the surface charges remain even after the illumination is switched off because their discharge mainly occurs through the solvent. Especially, we found that for our experiments with Novec 7500 ($\sigma = 4.46 \times 10^{-9}$ S cm^{-1}), that most of the particles were still tightly confined even 12 min after switching off the laser light. If necessary for special applications, the charge pattern on the surface can be periodically refreshed with very short laser illumination times.

FIG. 3. Electrophoretic trapping on a z-cut lithium niobate crystal; the inset shows the desired amplitude-modulated image honoring Bohr's discovery of the shell model in 1913; the microscope image shows that charged particles adhere well to the previously illuminated regions without any anisotropy.

In contrast to Novec 7500, in another related substance with a similar chemical structure, Novec 7300 (hydrofluorether of different chemical constitution, 3M), it was found that suspended GC particles in this medium are almost only of positive charge. This results in a different behaviour for both crystal orientations: On the −c face, particles are repelled from the illuminated regions, whereas on the +c face, the negatively charged illuminated regions attract positively charged matter. The experiment that proves this concept is illustrated in Fig. 4, which shows an exemplary two-dimensional illumination pattern and the resulting particle adhesion. In both cases, the crystal surface was illuminated for 5 s with a mean intensity of 70 mW cm^{-2}, one time with the +c face up ((a) and (b)) and the second time with the −c face up ((c) and (d)). The results clearly show that in the first case, GC particles suspended in Novec 7300 adhere to the bright regions in the pattern due to the negative charge accumulated here. In the other case, with the −c face up, they are repelled from the spots of high intensity areas in the pattern. A direct implication is that the knowledge about the crystal orientation allows the determination of the sign of charges based upon their trapping position. This behaviour deviates clearly from the previously demonstrated DEP trapping experiments, where even uncharged particles were captured by high field gradients.[5,10]

In conclusion, we have demonstrated a very simple all-optical method for the generation of arbitrary two-dimensional adhesive or repulsive patterns on a lithium niobate z-cut crystal. The generation of charges on the crystal surface is facilitated by the bulk internal photovoltaic effect, hence requiring no external electrodes. Using a y-cut

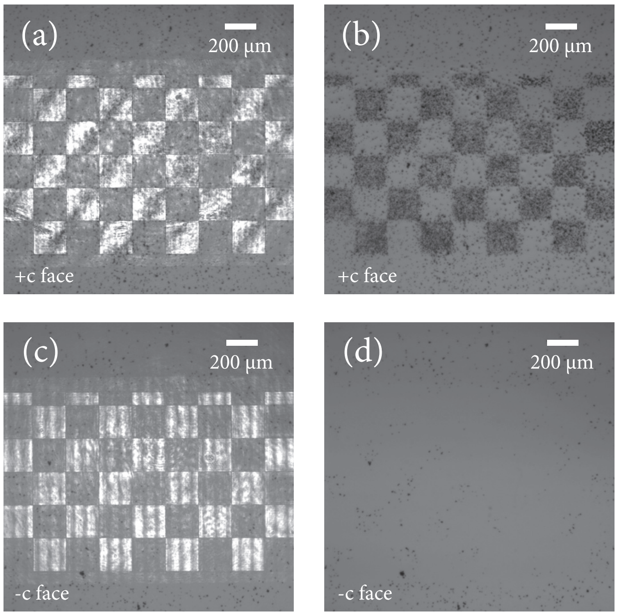

FIG. 4. Z-cut $LiNbO_3$ as charge sensor: (a) illumination of +c face with a binary square pattern and subsequent adhesion (b) of positively charged graphite spheres suspended in Novec 7300 to the illuminated regions; (c) illumination of −c face with the same pattern resulting (d) repulsion of positively charged particles from the high intensity regions.

lithium niobate crystal, it could be verified that particles suspended in an engineered fluid are indeed charged. This occurrence of charged particles in an otherwise uncharged solution and especially the different behaviour for the two related fluids Novec 7500 and 7300 is still subject to further investigation. Using the knowledge about the charged particles, the origin of the forces on a z-cut lithium niobate crystal can be described as purely electrophoretic. Since the charge transfer in lithium niobate is along the c-axis, the trapping scenario is inherently radial symmetric and not limited to the occurrence of field gradients, hence the resulting forces can be used to modify large areas without any precautions for the crystal orientation. Our approach is highly scalable from large scale applications on crystals in the cm range, to the small scale, with the pattern size physically restricted not by the substrate material but only by the size of the optical pattern that can be created, for example, by advanced techniques such as Talbot imaging. Due to the high optical quality and chemical inertness of the crystal samples, the technique lends itself to an easy integration in micro- and optofluidic surroundings such as Lab-On-A-Chip devices. Possible applications could include, for example, a fast method to generate two-dimensional arrays of special microparticles that can afterwards be transferred via moulding or printing. Additionally, the charges generated on the surface depend on the orientation of the crystals c-face which facilitates the use of a z-cut lithium niobate crystal as microfluidic charge sensor.

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