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Influence of the inhomogeneous field at the tip on quantitative piezoresponse force microscopy

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ABSTRACT Ferroelectric domain imaging with piezoresponse force microscopy (PFM) relies on the converse piezoelectric effect: a voltage applied to the sample leads to electromechanical deformations. In the case of PFM one electrode is realized by a tip, therefore generating a strongly inhomogeneous electric field distribution inside the sample which reaches values up to 10^8 V/m directly underneath the apex of the tip. Although often assumed, this high electric field does not lead to an enhancement of the electromechanical deformation of the sample. On the contrary, internal clamping of the material reduces the deformation as compared to the theoretically expected value which depends only on the voltage applied to the tip, thus being independent of the exact field distribution.

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1 Introduction

Since the first controlled switching of ferroelectric domains in lithium niobate (LiNbO_3) [1] a series of applications have been developed that require controlled structuring of domain patterns in the micron range. Besides the well known quasi-phase-matched frequency converters [2] and electro-optic scanners [3], nonlinear photonic crystals [4] and ultra-high density data storage devices [5] are intensively investigated. For further improvement of the domain patterns their visualization with a high lateral resolution is indispensable. Although several techniques have been developed [6], piezoresponse force microscopy (PFM) has become a standard technique in recent years mainly because of its very high resolution and ease of use [7].

Interestingly, quantitative data from PFM measurements are published very rarely. This deficiency is often justified by the presumption that the high electric field at the very tip would lead to an enhancement of the values measured, however, because the tip geometry and thus the exact field distribution is not known precisely a quantitative analysis of the data is not possible. Arguing along this way ignores the fact that, at least in a first approximation, it is not the electric field distribution but the applied voltage which determines the piezoelectric deformation of the sample. We carried out

experiments with different single-domain crystals, comparing the deformation underneath the tip with and without an additional top electrode. The measured values were found to differ seriously: using the tip as an electrode results in smaller deformations of the sample (sic!) by a factor of three as compared to those obtained with a large top electrode.

For a piezoelectric material of thickness t exposed to an external electric field E the (longitudinal) converse piezoelectric effect gives rise to a thickness change Δt proportional to the appropriate piezoelectric coefficient d :

$$\frac{\Delta t}{t} = Ed \Rightarrow \Delta t = Ud. \quad (1)$$

Note that the thickness change Δt does not depend on the thickness t of the sample but only on the applied voltage U . That is why for piezoelectric transducer elements, no thick crystals but stacks of multiple piezoelectric discs, all of them connected individually, are utilized [8].

In the case of PFM the electric field inside the sample is generated by the tip and a large back electrode. Typical dimensions are: tip radius $r \leq 100$ nm, thickness of the crystal is several $100 \mu\text{m}$, and the area of the back electrode is a few mm^2 . The voltage applied between the tip and back electrode usually is $10\text{--}20$ V which leads to an electric field strength $E = U/r \geq 10^8$ V/m at the apex of the tip. We assume a coordinate system with the z -axis perpendicular to the surface of the sample (thus z lies in the symmetry axis of the tip). Because the electric field E has a radial symmetry, electromechanical deformations due to field components perpendicular to z cancel out at the position of the tip apex. The electric field relevant for PFM measurements can therefore be reduced to its component E_z . Note that (1) is also valid if E_z is not homogeneous along the sample thickness t because $\int_0^t E_z ds = U$.

The piezoresponse force microscope is usually operated with an alternating voltage of $10\text{--}100$ kHz applied to the tip. In ferroelectric and thus piezoelectric samples this voltage causes thickness changes and therefore vibrations of the surface which lead to oscillations of the cantilever that can be read out with a lock-in amplifier. The generally observed frequency dependence of these measurements [9–11] was recently found to be independent of the kind of sample used. It is explained via a system-inherent background generated by the PFM setup [12].

However, when using a homogeneous top electrode to apply the alternating voltage, the situation is completely different: the whole sample and not only a restricted volume at

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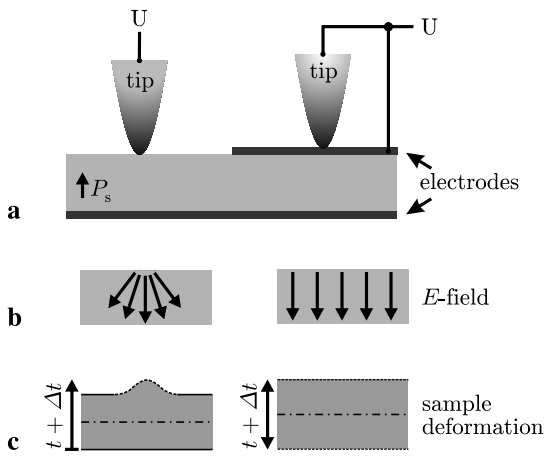


FIGURE 1 (a) Influence of the electrode configuration on the piezoelectric deformation. (b) Electric field distribution inside the crystal. (c) Resulting piezoelectric deformation of the sample. P_s : spontaneous polarization, U : applied voltage, t : sample thickness, Δt : thickness change

the very surface is vibrating, thereby also exciting the sample holder. In this case the frequency dependence is no longer independent neither on the sample nor on its mounting. Figure 1 schematically depicts the electric fields (b) and the expected deformations (c) for both electrode configurations (a). The extra wire connecting the top electrode is mandatory since an electrical contact via the tip can generally not be assured [13].

Quantitative and thus comparable values for the piezoelectric deformation with and without a top electrode can therefore be obtained only when overcoming this difficulty of different frequency dependencies for the two electrode configurations. For this reason we carried out the measurements presented in this contribution at very low frequencies (a few Hz) thereby avoiding any mechanical excitation of the PFM head, the sample, and sample holder. Below 10 Hz we did not observe any frequency dependence. We also confirmed identical piezoelectric deformations, both for the high (> 10 kHz) and the low (< 10 Hz) frequency regime when using the tip as an electrode. Note that image acquisition at low frequencies is not possible because a single data point requires an integration time up to several minutes.

2 Results and discussion

We investigated two single crystal samples (LiNbO₃ and α -quartz), each having a size of 10×10 mm², one side being half, and the other fully covered with a 100 nm thick conductive gold layer. For comparison we also acquired data using a periodically poled potassium titanyl phosphate (KTP) crystal [14]. For the determination of the piezoelectric deformation with the large top electrode, we positioned the tip in its center to avoid any edge effects of the gold layer. In the case of the tip acting as an electrode, we positioned it as far as possible away from the (now floating) gold layer to minimize the influence of a modified electrical field geometry. The measurements were carried out with a stand-alone scanning force microscope (SMENA, NT-MDT) modified to allow the application of voltages (here: 10 V_{pp} @ 5.2 Hz) to the tip. The lock-in amplifier (SRS 830) was set to 10 s integration time and each data point was averaged over half an hour.

Thereby we get sub-picometer accuracy even with a noise level of ≈ 0.2 pm/V present in our setup. For quantitative experiments every cantilever has to be calibrated individually, hence, we utilized one single cantilever (Micromasch) for the comparative measurements.

Table 1 summarizes the experimental results. The measurement on LiNbO₃ was repeated with different tips, hence we obtained slightly different values, put as an error. As our KTP is a multi-domain sample it is not possible to obtain meaningful data with a large top electrode (see also [13]).

The table also quotes the values of the appropriate piezoelectric coefficients found in the literature: for d_{11} of α -quartz there is a nearly perfect agreement in the publications. For LiNbO₃, however, the values reported for the d_{33} range are between 16–23 pm/V and 6–9 pm/V. Interestingly, for the other piezoelectric coefficients d_{15} , d_{22} , and d_{31} there is a reasonable good consensus in those publications. For KTP the values for d_{33} vary strongly as well.

On α -quartz, we find perfect agreement between the established value for d_{11} and our result with the large top electrode. The value determined with the tip acting as an electrode, however, is smaller by a factor of three. The same situation is also found for LiNbO₃ when referring to the large values for d_{33} reported in the literature. Similarly, the measurements on KTP are consistent with the observation that using the tip as an electrode leads to values of the piezoelectric coefficient that are too small by roughly a factor of three.

So why are the values of the piezoelectric coefficients obtained with the tip acting as an electrode too small? We believe that internal clamping of the sample constrains the mechanical deformation. Because of the inhomogeneity of the electric field, the full deformation must occur in a very small volume (μ m-sized). This, however, leads to clamping and thus to a reduced mechanical deformation. The load of the tip can not cause this effect, because the crystal undergoes the same load underneath the tip with a large an electrode. Increasing the load by a factor of 100 did not affect the values measured.

To underline our statement that the stiffness of the material itself limits the mechanical deformation we carried out measurements on a LiNbO₃ crystal with one single domain boundary, the backside being fully covered, and the front half-covered, with a gold electrode (Fig. 2). To obtain a measure of the stiffness of the material we performed line scans of 300 μ m across the domain boundary, as shown in Fig. 2. The measurement with the large top electrode leads to a full width at half maximum of the slope at the domain boundary of ≈ 100 μ m. In this configuration, clamping takes place at the domain boundary which itself is not excited, because of the

Sample	PFM-tip (pm/V)	PFM-top (pm/V)	Literature (pm/V)	[Ref.]
LiNbO ₃	6 ± 1	20 ± 2	16–23 6–9	[15–17] [18–21]
KTP	6.1	–	10–26	[22–24]
α -quartz	0.8	2.3	2.3	[25]

TABLE 1 Comparison of the piezoelectric deformation measured with PFM (tip as the electrode or an extra top electrode) for three different samples (d_{33} for LiNbO₃, d_{33} for KTP, and d_{11} for α -quartz). In addition, values from the literature with corresponding references are given

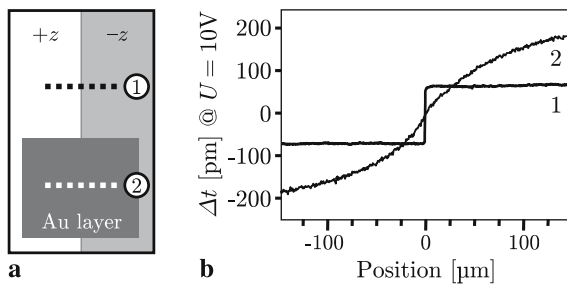


FIGURE 2 Schematics of the LiNbO₃-sample used for the PFM measurements. ① and ② indicate the positions of the line scans. (b) Line scans recorded at the positions indicated in (a) across the domain boundary situated at position 0

symmetric domain configuration. This can be compared to the situation where the tip acts as an electrode and at a distance of some microns from the tip apex the electric field has decreased to $\ll 1\%$ of its initial value [26]. The stiffness of the material, however, does not permit a strong deformation within some microns, therefore the required electromechanical response of the material is restricted. For comparison we also show the line scan obtained with the tip acting as an electrode. The domain boundary appears as a sharp edge with a full width at half maximum in the order of the tip radius [27]. The amplitude, however, is smaller by a factor of three as compared to the measurement with the large electrode.

The strongly inhomogeneous field might also lead to another effect reducing the deformation of the crystal: it is not ensured that the material still responds linearly on applying an electric field of 10^8 V/m, as it is present at the very tip. This would also lead to values of the piezoelectric coefficient being smaller than expected. From the results presented in the last paragraph, however, this effect is not dominant.

A further point is: why are the values measured with the tip as an electrode too small by roughly the same factor for all samples? We explain this by the stiffness of the materials which is of the same order of magnitude [19, 23, 25], the samples thereby undergo similar mechanical clamping. Performing experiments with single crystals exhibiting very different elastic properties should therefore lead to other factors. Ceramics, however, behave completely different, they are much softer than single crystals. Therefore comparative measurements have not shown a tip effect on their piezoelectric deformation [28].

As a result from our measurements, quantitative determination of piezoelectric coefficients with the PFM technique can not be achieved by simply putting the sample to be measured into the experimental setup. In addition to the system inherent background at high frequencies [12], that can be more or less circumvented by measuring at very low frequencies, one has to account for the tip effect as analyzed in this contribution. Therefore a calibration with a sample of known piezoelectric coefficient (e.g. α -quartz) holds as many experimental traps as when using PFM to determine piezoelectric coefficients [29]. This might be the reason why, despite its potential to achieve ultra high vertical resolution (sub-picometer), PFM measurements yielded only reasonable piezoelectric coefficients.

Note: calculations like the one in the paper by Kolosov et al. [30] are simply wrong: admittedly, the electric field at the very tip can reach values up to $E_{\text{tip}} = 2.5 \times 10^7$ V/m, how-

ever, this electric field does not persist along the whole thickness of the crystal. Therefore the value of $\Delta t = 2tE_{\text{tip}}d_{33} = 30$ nm (sample thickness $t = 0.3$ mm, piezoelectric coefficient $d_{33} = 2 \times 10^{-12}$ m/V) is incorrect.

3 Conclusion

In this contributions we tried to clarify, despite its simplicity, an apparently still not generally accepted feature of PFM imaging: the surface deformation of the sample is not enhanced due to the high electric field at the very tip. Instead, the inhomogeneity of the field leads to internal clamping and thus a reduced electromechanical deformation of the sample. With our experiments we hoped to have disposed of the idea of large PFM signals due to electric field enhancement by the tip. This is not the case, neither from a theoretical point of view nor from experimental results. On the contrary, the values measured with PFM are smaller than expected.

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