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Non-piezoelectric effects in piezoresponse force microscopy

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ABSTRACT

Piezoresponse force microscopy (PFM) has been used extensively for exploring nanoscale ferro/piezoelectric phenomena over the past two decades. The imaging mechanism of PFM is based on the detection of the electromechanical (EM) response induced by the inverse piezoelectric effect through the cantilever dynamics of an atomic force microscopy. However, several non-piezoelectric effects can induce additional contributions to the EM response, which often lead to a misinterpretation of the measured PFM response. This review aims to summarize the non-piezoelectric origins of the EM response that impair the interpretation of PFM measurements. We primarily discuss two major non-piezoelectric origins, namely, the electrostatic effect and electrochemical strain. Several approaches for differentiating the ferroelectric contribution from the EM response are also discussed. The review suggests a fundamental guideline for the proper utilization of the PFM technique, as well as for achieving a reasonable interpretation of observed PFM responses.

1. Introduction

The increasing demand for miniaturized electronic devices has prompted the development of novel techniques for characterizing material properties accurately at the nanoscale [1,2]. In this perspective, scanning probe microscopy (SPM) has enabled new approaches for evaluating nanoscale material properties. Variants of SPM have been developed and extensively utilized to explore elastic [3–5], electrical [6–9], electrochemical [10–12], magnetic [13–16], and various functional properties [17–20] in the last few decades. Among them, piezoresponse force microscopy (PFM) has been particularly useful for exploring ferro/piezoelectric properties, owing to its remarkably high resolution and ease of use [21–24]. To date, the capability of PFM for probing nanoscale ferro/piezoelectric phenomena has been extensively demonstrated in ferroelectric oxide systems [25]. The scope of PFM applications has recently expanded into the fields of polymeric [26–28], biological [29–31], and organic–inorganic hybrid materials [32–35], and also to two-dimensional materials [36–38], with a view to investigating ferro/piezoelectricity. However, owing to the intrinsic imaging mechanism of PFM, several artifacts related with topographical features [39,40] and background signals [41–43] can affect PFM measurements. Apart from these artifacts, there are concurrent non-piezoelectric effects, which contribute directly to the PFM response. In other words, the non-piezoelectric effects can cause an additional electromehcanical (EM) response that can be simultaneously detected as a PFM response and thereby distort the PFM measurement. This problem strongly motivates an understanding of the operational principle underlying PFM and of non-piezoelectric effects, to explicitly interpret PFM measurements.

1.1. Principle of PFM

Ferroelectric materials display a spontaneous polarization that can be switched by the application of an external electric field. They also possess piezoelectric behavior [44,45], described as a reversible linear relation between a mechanical deformation and an electric field. In general, the process of mechanical deformation, *i.e.* surface volume change (an expansion or contraction), induced by an electric field is referred to as the inverse piezoelectric effect in a ferro/piezoelectric material. In PFM, a periodic surface volume change can be induced by applying an ac voltage to the conductive tip of an atomic force microscope (AFM), then detecting the EM response through the AFM cantilever [46]. This particular EM response constitutes the PFM response and is quantified in terms of its amplitude and phase, by comparing the input ac voltage with periodic surface vibrations (the output signal from the sample) using a lock-in amplifier, as outlined in Fig. 1(a) [21]. In general, the PFM amplitude (A) and phase (φ) signals characterize the magnitude of the piezoelectricity and the polarization direction, respectively.

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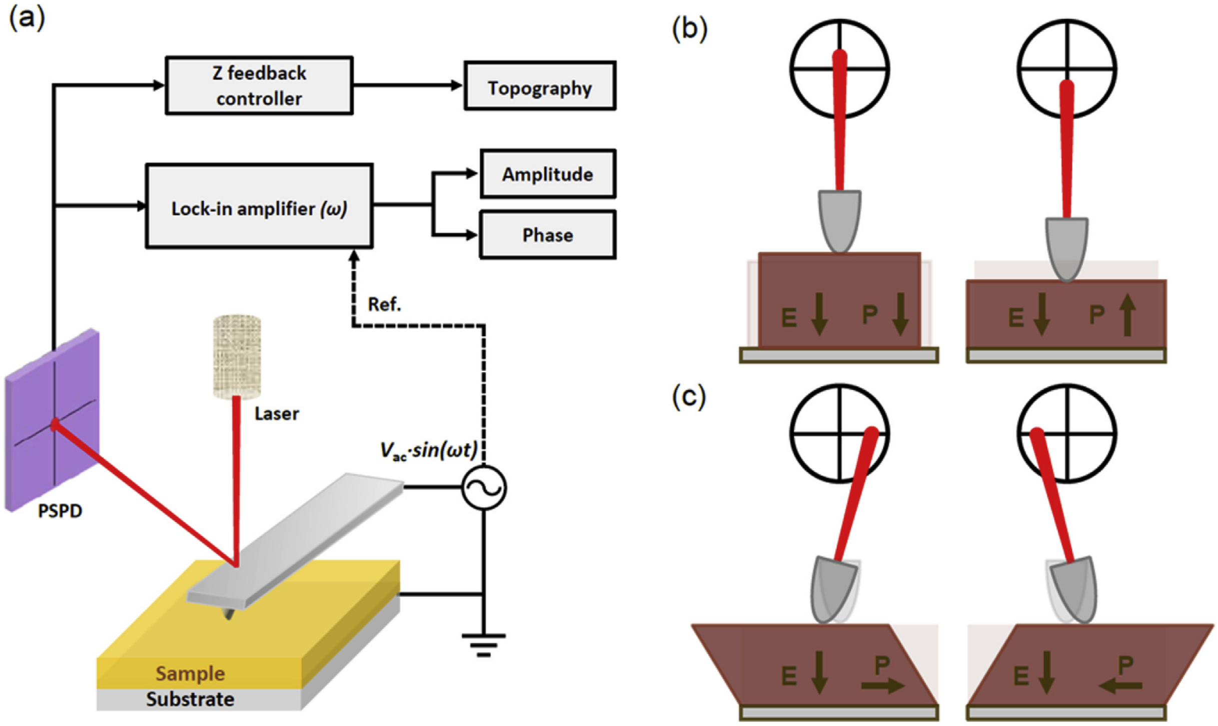

Fig. 1. (a) Schematic PFM setup, and the principle of (b) vertical and (c) lateral PFMs.

Together, they make up the piezoresponse $A\sin(\varphi)$. Analyzing the vertical (lateral) cantilever deflections, as shown in Fig. 1(b) and (c), yields information on the vertical (lateral) component of the polarization, an approach referred to as vertical (lateral) PFM [47,48]. On the basis of the operational mechanism, vertical and lateral PFMs can provide profound insight into nanoscale ferro/piezoelectric phenomena such as piezoelectricity [49–52], polarization switching dynamics [53–56], domain growth, and wall motions [57–60]. Three-dimensional domain structures can also be explored by combining vertical and lateral PFMs [61–65].

The observed PFM response is generally expressed as

$$(PR)_\omega = d_{eff} V_{ac} \sin(\omega t) \tag{1}$$

where d_{eff}, V_{ac}, ω, and t are the effective piezoelectric coefficient, magnitude and frequency of the ac voltage, and time, respectively. The observed response by PFM is thus directly governed by piezoelectric properties such as the piezoelectric tensor of ferro/piezoelectric materials. A conventional PFM measurement is performed by applying a single ac frequency that is generally far from the contact-resonance frequency of the cantilever. However, resonance-enhancement techniques based on the use of the contact resonance frequency have been suggested. These techniques, which include dual ac resonance tracking (DART) [66,67] and band excitation (BE) [68,69], significantly enhance the signal-to-noise ratio of the observed PFM response, which facilitates the achievement of vertical (z-height) resolutions at the picometer level. Such techniques have been used extensively for PFM imaging and spectroscopic approaches, *e.g.*, switching spectroscopy PFM [70], to explore local ferro/piezoelectric properties at the nanoscale [71,72].

1.2. Origins of the EM response in PFM

Although the piezoresponse of ferro/piezoelectric materials is of interest as the main origin of the induced EM response in the PFM, several other non-piezoelectric effects can also contribute to the EM response, as illustrated in Fig. 2. These non-piezoelectric effects not only hinder accurate PFM measurements but also, in some cases, cumulatively dominate the measured PFM response [73–75]. It is therefore very important to acknowledge and understand their influence to allow an accurate interpretation of material properties measured by PFM.

This review primarily discusses two major non-piezoelectric effects that directly affect the interpretation of PFM results: the electrostatic effect and electrochemical strain. Section 2 addresses

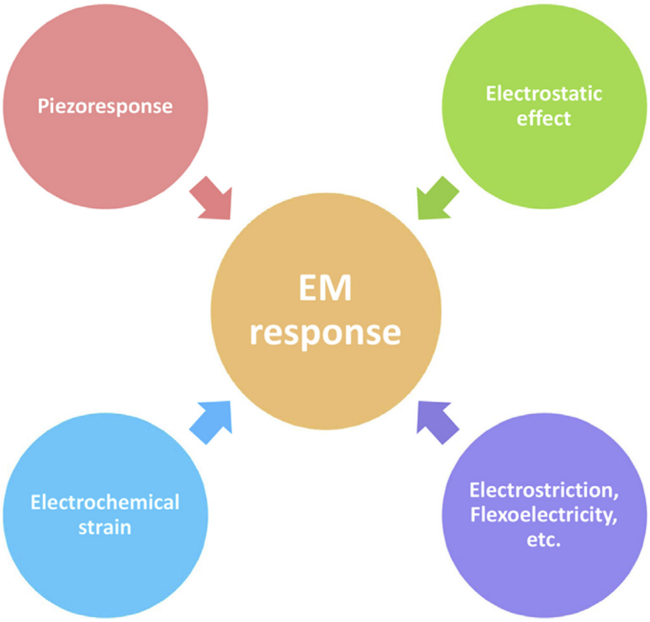

Fig. 2. Schematic diagram of origins that can induce EM response.