Polarization behavior of ferroelectric multilayered composite structures

Y. T. Or and C. K. Wong[a]

Department of Applied Physics, The Hong Kong Polytechnic University, Hong Kong,
People's Republic of China

B. Ploss

Fachhochschule Jena, FB Physikalische Technik, University of Applied Sciences, Carl–Zeiss-Promenade 2,
07745 Jena, Germany

F. G. Shin

Department of Applied Physics, Materials Research Center and Center for Smart Materials,
The Hong Kong Polytechnic University, Hong Kong, People's Republic of China

We model the polarization behavior of ferroelectric multilayered composite structures including the double layer and the triple layer under the action of variable fields. The model takes into consideration the ceramic/polymer interfacial charge and the nonlinear hysteresis nature of the ferroelectric constituent materials to study the poling process of multilayered structures. The results obtained are compared with available experimental results on double-layer lead zirconate titanate/polyvinylidene fluoride-trifluoroethylene and triple-layered triglycine sulphate/polyvinylidene fluoride-trifluoroethylene/triglycine sulphate systems. In general the broad features of the experimental results were reproduced, and fairly good agreement between the predictions and the experimental results was found. The work also demonstrates that electrical conductivity in the ferroelectric materials is an important factor controlling the poling process. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1558961]

I. INTRODUCTION

Composite systems of ferroelectric ceramics and ferroelectric polymers combine the high pyroelectric and piezoelectric activities of the ceramic with the excellent mechanical properties of the polymer, thus deriving tailor-made advantages for sensor and transducer applications. Ferroelectric materials possess spontaneous polarization but usually show no net macroscopic polarization due to the different orientations of the individual domains. In order to impart piezoelectric and pyroelectric activities, they must be subjected to a poling process, to align the spontaneous polarization in the material(s). In a ferroelectric ceramic/polymer composite, the permittivity of the ferroelectric ceramic is normally much higher than that of the polymer, thus the electric field acting on the ceramic phase would be much lower than the applied electric field.[1] In this context, it seems impossible to pole the ceramic in the composite. However, successful thermal poling of various composite systems like lead titanate, lead zirconate titanate (PZT), and lanthanum modified lead zirconate titanate with polymer have been reported in the literature.[2–4] However, the poling recipes/techniques are still quite empirical because a firm understanding of the physical processes involved in poling has not been fully established.

The present work aims to systematically analyze the physical process(es) pertaining to the poling of the multilayered composite. Understanding of the physical process behind poling will give an idea on the materials selection and the poling parameters for particular applications. In this article, we shall first give a theoretical description on poling of multilayered composite systems by considering the continuity of the total current density going through each ferroelectric layer along the poling direction. A modified model of Miller *et al.*[5–7] is used for the description of the $P-E$ relation of each ferroelectric layer. The modeling will be applied to study the switching of the bilayer PZT/polyvinylidene fluoride-trifluoroethylene P(VDF–TrFE) composite investigated by Furukawa *et al.*[1] and the poling of P(VDF–TrFE) with ferroelectric triglycine sulphate (TGS) electrodes investigated by Ploss *et al.*[8] Then we shall show the comparison between the experimental results with the theoretical predictions.

II. THEORY

Consider a multilayered composite consisting of n ferroelectric layers connected in series along the thickness direction (see Fig. 1). The constitutive equation of the ith layer is given by

$$D_i = \varepsilon_i E_i + P_i, \tag{1}$$

where i is an integer between 1 to n. D, ε, E, and P represent the dielectric displacement, permittivity, electric field, and ferroelectric polarization, respectively. P_i generally depends on E_i in a complicated manner and a model for describing $P-E$ relations must be used, as will be explained later. In contrast, when some layer, say jth layer, is nonferroelectric, then $P_j = 0$.

a)Electronic mail: wongck.a@polyu.edu.hk