Ferroelectric Properties of Polymeric Bilayer Systems

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ABSTRACT

Bilayers of VDF-TrFE ferroelectric copolymer thin films have been characterised in terms of polarisation behaviour and dielectric nonlinearities. The effective properties of the ferroelectric bilayers have been investigated and compared with those of the individual layers in the stack. It has been demonstrated that the effective permittivity of the bilayer is not fully described by a simple series model. The remanent polarisation P_r in the bilayer shows essentially a linear decrease with temperature and reaches zero at around 120 °C, i.e. above the Curie temperatures of the two copolymers involved. The two observations are attributed to electric bias fields in the bilayer which cause a coupling between the two layers and lead to a temperature dependence of the polarisation which is phenomenologically similar to observations in superlattices.

Index Terms — **Nonlinear media, ferroelectric films, plastics, nonhomogeneous media, dielectric polarization.**

1 INTRODUCTION

MULTILAYER structures of ferroelectrics and dielectrics have attracted a great interest during the last decade. While ceramic systems have been extensively studied, polymeric systems were left untouched. Due to its unique properties the ferroelectric copolymer VDF-TrFE is of great interest, e.g. a promising candidate for ferroelectric memory applications [1]. The formation of bilayers and multilayers is a way to modify the dielectric and polarisation properties and optimise them for the needs of applications. We want to extend the knowledge gained on single layer properties to multilayers [2].

2 PHENOMENOLOGY

In the following section a model is derived for the free energy of a stack of n layers including the $n-1$ interface layers appearing inevitably. The metal-dielectric interface introduced by the deposition of electrodes is not part of the discussion, knowing that for a stack with a small number of thin layers it may become relevant.

2.1 FREE ENERGY OF A FERROELECTRIC MULTILAYER

A common approach to describe ferroelectric materials in the vicinity of a transition from a prototype phase of high symmetry to a ferroelectric phase of lower symmetry is to introduce a parameter reflecting the appearance of an order. In order to stick to the principle of free energy which includes external properties, e.g. the Maxwell electric field E, the electric displacement D is the proper order parameter.

Furthermore, it is common to express the thermodynamic potential as a deviation from the prototype phase $F = F_{FE} - F_{PT}$ were F_{FE} and F_{PT} are the free energies of the ferroelectric phase and the prototype phase, respectively. Since we describe a system containing two types of ferroelectric materials owing at least two different critical temperatures, we include the energy of the prototype phase in our theoretical considerations. To further simplify our model we assume zero stress and a uniformly distributed polarisation within each layer. Latter can be achieved by using the free energy density f with $F = \int f(T, D)dV$. The free energy of a system of n layers possessing $m = n - 1$ interfaces can then be expressed as:

$$F = \sum_{i=1}^{n} \left[\left(\frac{\alpha_i}{2} D_i^2 + \frac{\gamma_i}{4} D_i^4 + \frac{\delta_i}{6} D_i^6 + f_{PT,i} \right) - E_i D_i \right] V_i + \sum_{j=1}^{m} \left(f_{I,j} - E_{I,j} D_{I,j} \right) V_{I,j} \tag{1}$$

Manuscript received on 7 September 2014, in final form 11 April 2015, accepted 5 August 2015.

DOI 10.1109/TDEI.2015.004909