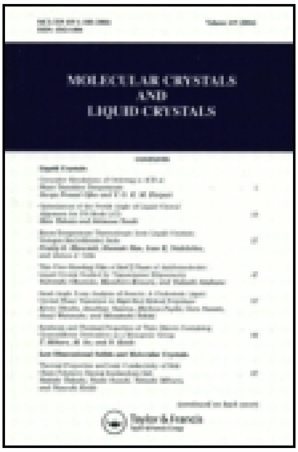

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Sergei Fedosov [a]
[a] Odessa Technological Institute, Odessa, Ukraine

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MOBILITY AND TRAPPING OF INJECTED CHARGE CARRIERS IN FERROELECTRIC ORGANIC SOLIDS

SERGEI FEDOSOV
Odessa Technological Institute, Odessa, Ukraine

Abstract A model is proposed and discussed explaining the role of trapped charge carriers in stabilization of polarized state in ferroelectric polymers.

INTRODUCTION

Ferroelectric organic solids such as poly(vinylidene fluoride) or PVDF are of grate practical importance because of their strong piezo- and pyroactivity, which are closely related to a high level of remanent polarization.
The three-stage model of polarization build-up in ferroelectric polymers was worked out in our previous studies based on experimental data on kinetics of the electret potential, absorption currents, voltage-current characteristics and thermostimulated polarization currents.[1-4]
It was shown that strong polarization appeared near a positive electrode at the second stage and its expantion was similar to the movement of a domain wall in conventional ferroelectrics. Charge transport equations were solved analytically assuming an important role of injected and deeply trapped charge carriers. By the curve fitting technique, it was found that the effective mobility of charge carriers decreased gradually reaching the value of 1.1×10^{-8} $m^2/V.s$. It corresponded most likely to the trap modulated mobility, since it was six orders of magnitude lower than that of free carriers. However, a mechanism of the charge trapping and its influence on the stability of polarization were not clearly understood.

CHARGE TRAPPING

It is obvious that charge trapping may occur not only at the permanent local centres but also at the potential wells related to large-scale fluctuations of the potential.
Let us consider the possibility of such a process in ferroelectric organic solids composed of ferroelectric crystallites (such as Form-1 of PVDF) dispersed in the amorphous polymer matrix. In case of the uniformly polarized spherical crystallites, potential energy of an elementary charge at its surface is defined by the interaction between the charge and a local field of the polarized crystal

$$E = -(1/3e_o)ePR\cos\alpha, \qquad (1)$$

where e_o is the permittivity of a vaccum, e the elementary charge, P the vector of polarization, R the radius-vector, α the angle between P and R.
It is clear from Eq. (1) that the energy is negative at half of the surface. Conditions are favourable there for localization of charge carriers.
Suppose n is the surface density of the localization sites and g(E) is the energy distribution function of the traps. Then one can write

$$g(E)dE = ndS, \qquad (2)$$

where $dS = 2\pi R^2\sin\alpha d\alpha$. (3)

It follows from Eqs. (1) to (3) that

$$g(E) = 3e_oN/ePR = \text{const}, \qquad (4)$$

where N is the total number of traps ($N = 2\pi R^2 n$). Therefore, in a range of 0 to $E_{min} = -ePR/3e_o$, according to Eq. (1), there is a uniform (rectangular) energy distribution of localized states at the surface of the polarized crystallites. For quantitative evaluation one can take $V = 10^{-19}$ cm^3 (R = 3o Å). Considering $P = 0.14$ C/m^2 according to [5] we obtain $E_{min} = -15$ eV, indicating

the presence of deep trapping of the charge carriers at the crystallite surface,
In case of crystallites having a form of plane disc (with h as the thickness) one can get

$$E = - hP\cos\alpha/4e_0. \qquad (5)$$

Substituting $h = 10$ Å and $P = 0.14$ C/m^2 we obtain $E_{min} = - 4$ eV, also indicating existance of deep trapping of the carriers.
Therefore, during the poling, new traps are added to the already existed Anderson's centres of localization in amorphous phase and to the Maxwell-Wagner's traps at the crystal-amorphous interface. As a result, a thernodynamic equilibrium between the delocalized and the trapped carriers moves in a direction of the latter. This must lead to a decrease in the effective mobility and conductivity while strong polarization is formed (the field exceeds a coercive value). These features were observed experimentally in our previous works[1-4], proving validity of the proposed model.

DISCUSSION AND CONCLUSION

If the vector P is directed oppositely under the influence of an external field, the energy state of the previously localized charge carriers will also be changed drastically. As a result, charge carriers will be shaked out the traps, while another traps are formed at the opposite side of the crystallite. Although the total number of traps and their energy distribution remain almost the same, their spatial distribution will be different.
A mass release of carriers from the traps can be considered as a peculiar internal emission. For a definite time, there is a transient period of abnormally high conductivity due to increase of the free carrier density. The phenomenon was observed in our previous works[1-4] supporting the proposed model.
According to the model, the trapped charges play an

important role in processes of stabilization and relaxation of the polarized state, since they partly compensate local depolarization fields.

One must distinguish between charge trapping in ferroelectric polymers and charge accumulation due to the Maxwell-Wagner's effect observed in all heterogeneous dielectrics. In the former case, the process is non-linear and irreversible. Moreover, strong polarization in crystallites and trapped charges at their surfaces form a stable self-balanced system even after the external field is removed. Considering the more or less universal approach developed in this work, we believe similar processes take place in composites containing ferroelectric inorganic crystals (such as PZT) dispersed in a polymer matrix.

In all cases, to obtain a high level of remanent polarization in ferroelectric polymers, it is necessary to provide, during the poling process, sufficiently high density of charge carriers either by their injection, for example from corona, or by heating as in the case of the thermostimulated polarization.

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