Small polaron hopping in $Fe:LiNbO_3$ from microscopic modelling to macroscopic observables

Laura Vittadello,[1,2] Mirco Imlau,[1,2] Marco Bazzan[3]
[1]Department of Physics, Osnabrück University, Barbarastraße 7, Osnabrück, Germany
[2]Research Center for Cellular Nanoanalytics Osnabrück (CellNanOs), Osnabrück University (Germany)
[3]Physics and Astronomy Department, University of Padova and INFN, Padova (Italy)

Iron doped lithium niobate is one of the most used ferroelectric oxides for photorefractive applications, such as the realization of high quality volume gratings, optical memories, holography and integrated circuits with all-optical capabilities [1]. Nowadays, for the same motivation, it starts to gain attention as ferroelectric photovoltaic material [3].

It is already accepted that the light-induced charge transport properties in this system can be understood to a large extend in terms of small polaron hopping.

In $Fe:LiNbO_3$ three different small electron polaron types are generally considered [2]: polarons localized on regular Nb sites, or free polarons (F); polarons localized on Nb_{Li} antisite defects (P); polarons localized on Fe_{Li} substitutional defects (Fe). Each of the former two can move by thermal assisted hopping among different sites, until a deep Fe trap is encountered.

Despite this advanced level of microscopic understanding, there is still a lack of knowledge on the relation between the macroscopic observable such as polaron mobility and lifetime with the basic polaron hopping processes. This missing knowledge is one of the main motivation preventing further tailoring of the mentioned applications. Here a first step in this reconciliation is proposed.

The Marcus- Holstein hopping model provides an explicit formula to compute the average hopping frequency $\omega_{i,j}$ between two different polaronic sites i and j at a given distance and for a given temperature. This formula lends itself to a numerical procedure based on a Monte Carlo approach to simulate macroscopic quantities as a result of the random motion of a statistically significant number of polaron hoppers [4]. It is possible to follow every polaron jump and keep track of its position, the time between the jumps and the different types of visited sites. These three basic information permit to reconstruct macroscopic quantities such as the polaron lifetime or the mobility, bridging the gap between the micro and the macro description.

In Fig. 1 a) a typical output is shown. In the upper part the green curve represents the survival probability of an antisite polaron created by photo-excitation. In the bottom of the figure the number of hops performed as P and F polaron are shown. The particular shape of the polaron decay is determined by the dominant polaron jumping mechanism at the different time scales [6]. At short times, it can be seen that the dominating polaron trapping mechanism (assumed to start initially on an antisite defect) involves fast direct hopping towards iron traps in one single jump, without percolation on other mobile sites. This first regime is named as *direct trapping regime*. In the long time scale, we encounter decay processes corresponding to polarons performing a certain number of hops before encountering a deep iron trap and end their life. The crossing point between the two regimes occurs at a time τ_0 corresponding to the typical time for the polaron to hop on a nearby transport site.

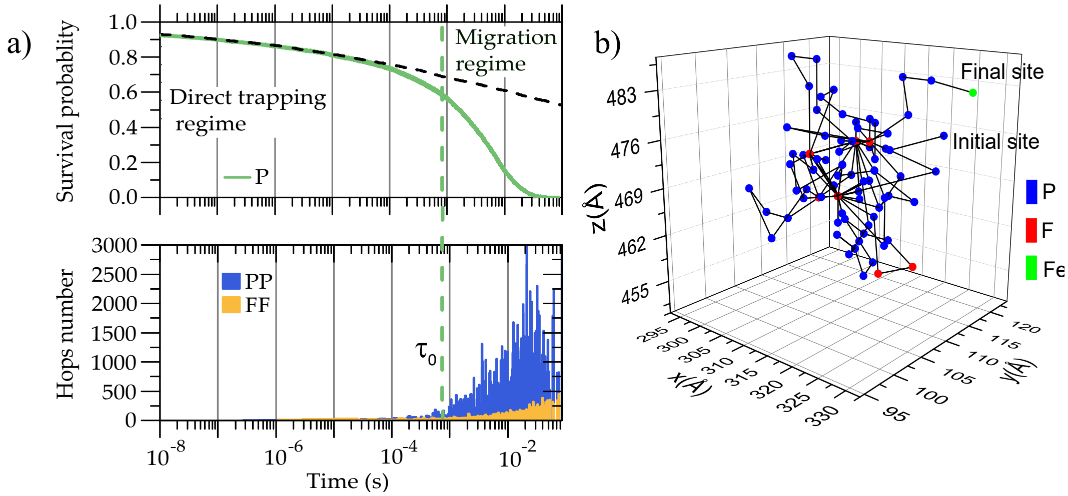

Figure 1: a) (Top) Survival probability of P polarons as measured by a typical LIA experiment. The black dashed line shows the decay shape when polaron migration is prohibited. (Bottom) Number of PP and FF hopping processes performed by each polaron during its walk.. b) Exemplary result of a polaron trajectory just after $t = \tau_0$.

After all the "quick" direct trapping processes have occurred, it becomes more probable that the polaron hops on a transport site rather than stay on its original site until a direct hop on a Fe trap takes place. An exemplary result of a polaron trajectory just after $t = \tau_0$ is shown in Fig. 1 b). This regime is indicated as *migration-accelerated regime*.

The second important macroscopic quantity that can be modelled in term of polaron hopping diffusion is the polaron mobility μ. Generally it is assumed that charge carriers are normally diffusing particles with a time independent mobility. This assumption is justified in the case of free polarons, since the distribution of the hopping times satisfies the hypothesis of the central limit theorem. However bound polarons are characterized by an heavy-tailed distribution of hopping times which lead to the so-called anomalous diffusion behaviour. In this context, mobility is a time-dependent quantity leading to unusual phenomenology for the macroscopic charge transport processes when the latter are dominated by bound polarons. Monte Carlo simulations are helpful also to investigate these regimes.

References

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