

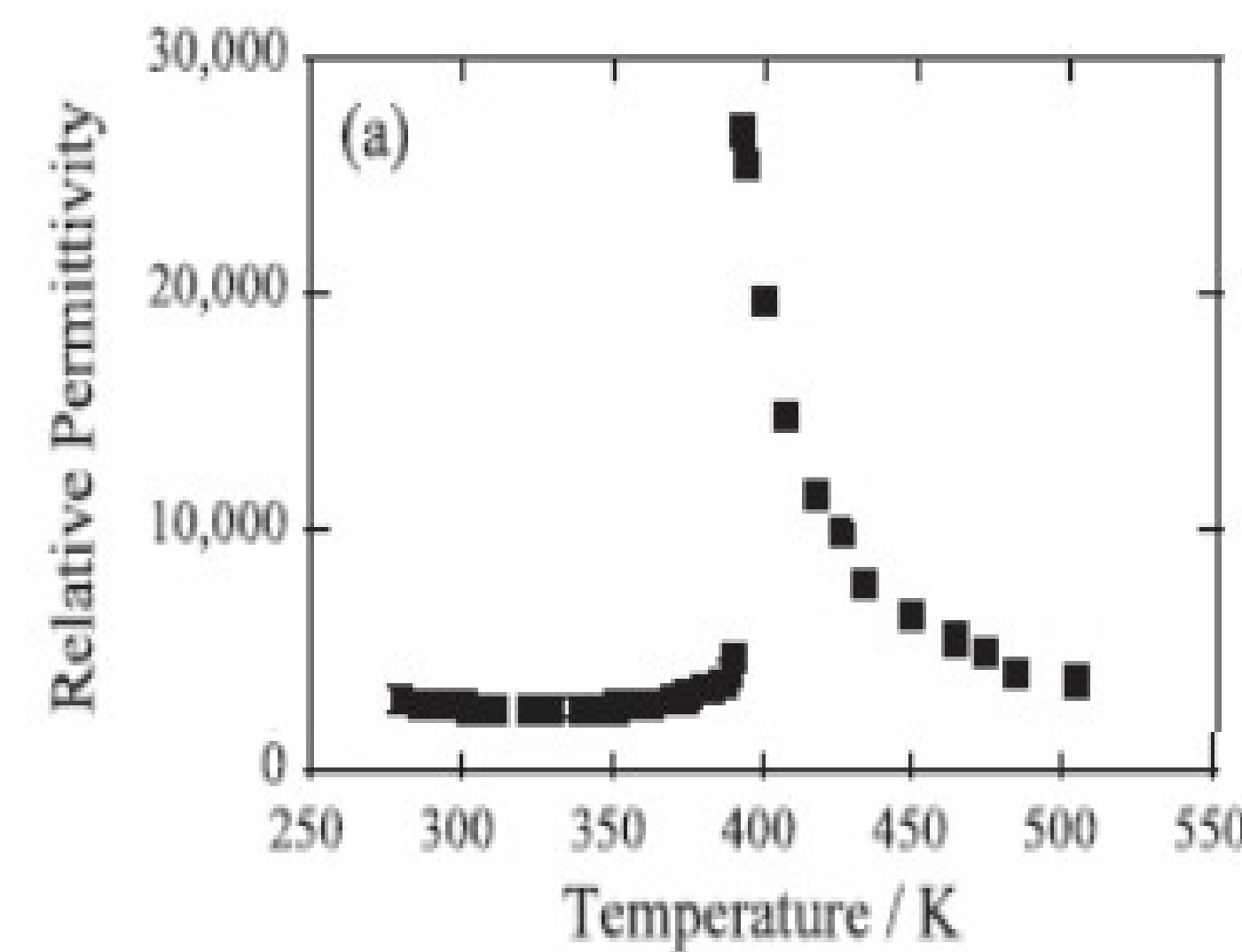
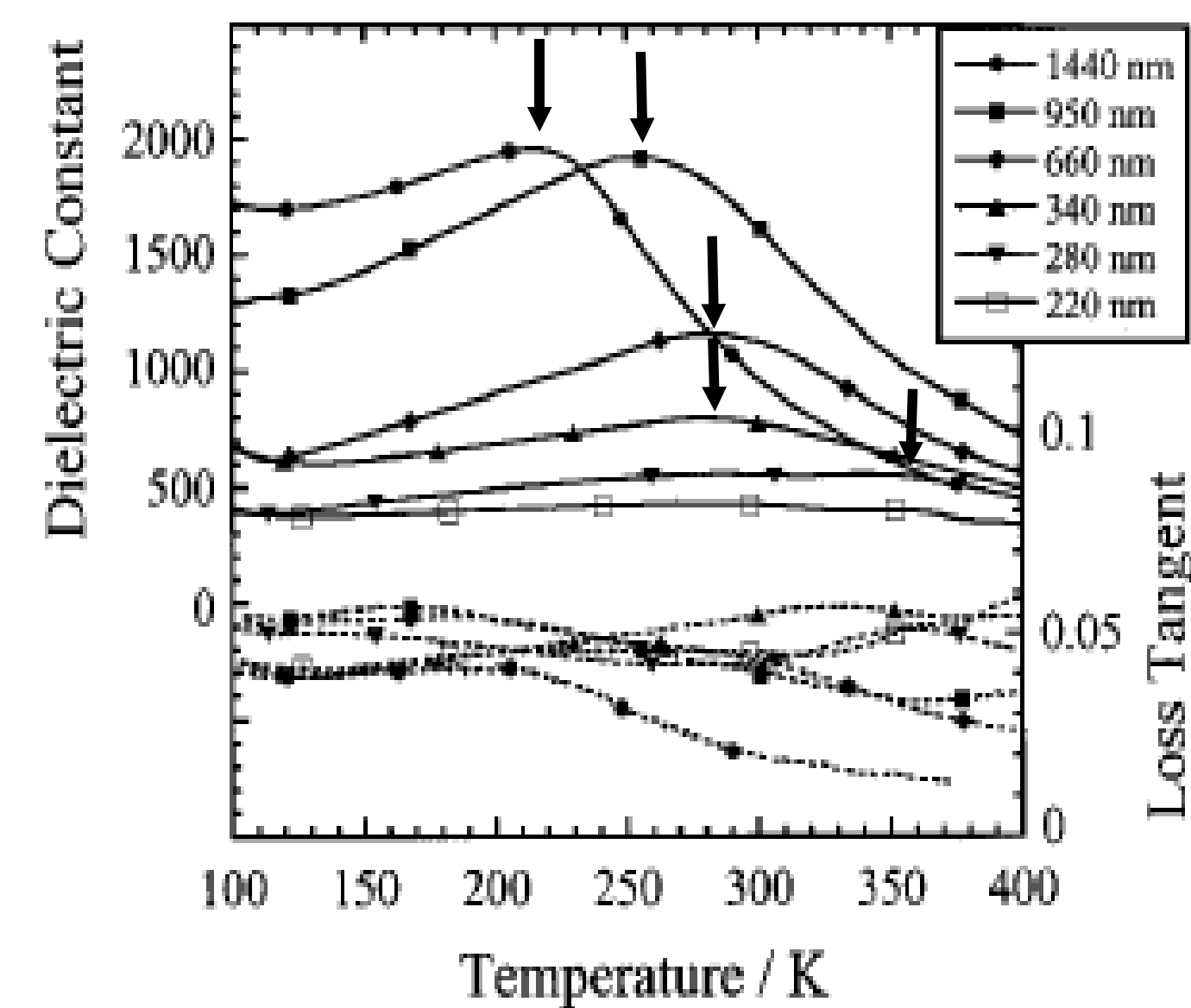
# Modeling Thickness-Dependence of Ferroelectric Thin Film Properties

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## Introduction

- Interest in **nanoscale** ferroelectrics; applications in nonvolatile and low-power consuming ferroelectric memories; demands for higher densities
  - Smaller scales: **Size-dependent effects** are important; dielectric properties in ferroelectrics depend on electromechanical BC due to the long-range nature of the underlying electrostatic interactions
  - Strong **coupling** between **polarization** and **strain** [e.g. Curie temperature is higher when the film is strained]
- Experimental motivation:
- Thin films grown on substrates show **broadened dielectric permittivity** ( $\epsilon_r$ ) near para-ferroelectric transition;  $\epsilon_r$  is more suppressed for thinner films (the temperature at which the maximum of the permittivity occurs is shifted to higher values for thinner films – arrows in the figure below)
  - Free-standing FE lamellae (75nm) yield bulk-type dielectric responses
  - Is **inhomogeneous strain necessary** to explain experiments **on planar films?**

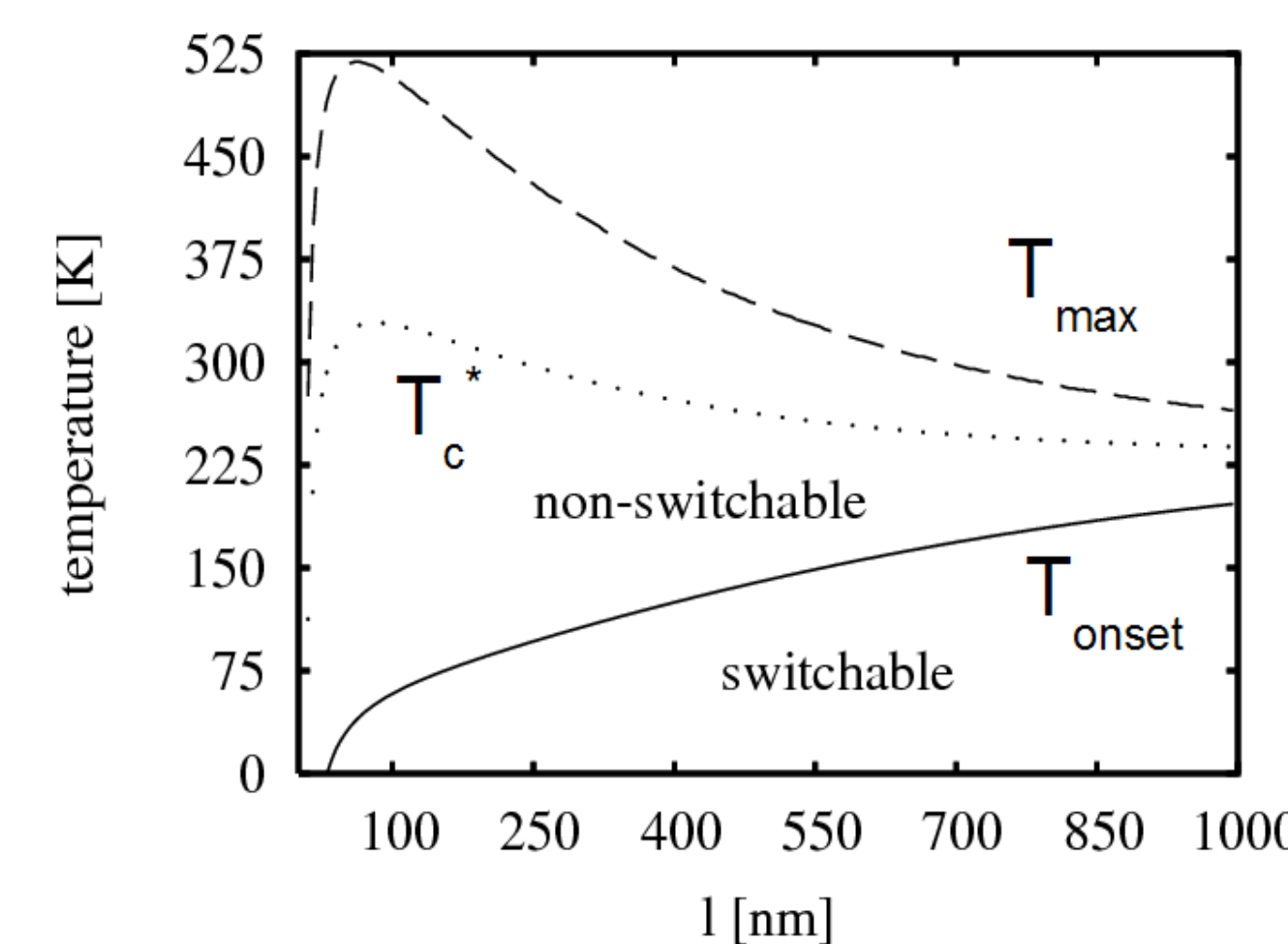


L.J. Sinnamon et al., Appl. Phys. Lett. **81**, 889 (2002) M.M. Saad et al., J. Phys.: Cond. Matt. **16**, L451 (2004)

## Landau-Ginzburg theory

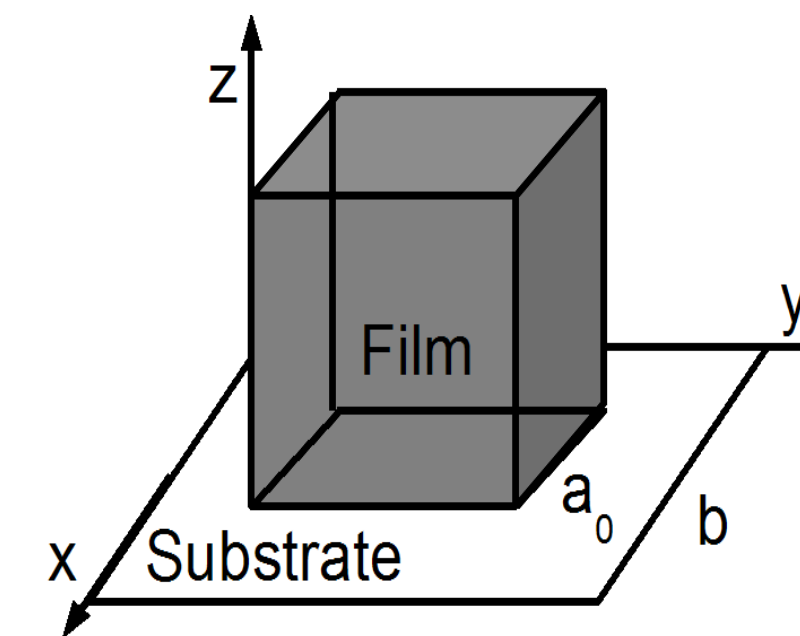
- B.C.  $\Rightarrow T_c^*(u_l), \gamma^*$ , depolarization field  $\Rightarrow T_c^*(l)$ , 'net' field =  $E_{ext} + W_l$
- $G_{LG}(T, P, u) = G_0 + 1/2 \beta(T - T_c^*(u))P^2 + 1/4 \gamma^* P^4 - (E_{ext} + W_l)P$
- Cubic equation for polarization:  $\beta(T - T_c^*)P_l + \gamma^* P_l^3 = E_{ext} + W_l$
- Susceptibility ( $\chi = \epsilon_r - 1$ ):  $\chi^{-1}/\epsilon_0 = \beta(T - T_c^*) + 3\gamma^* P_l^2$

- Separation of temperature scales: onset of reversible spontaneous polarization ( $T_{onset}$ ), theoretical Curie temperature ( $T_c^*$ ) and the maximum of dielectric permittivity ( $T_{max}$ )



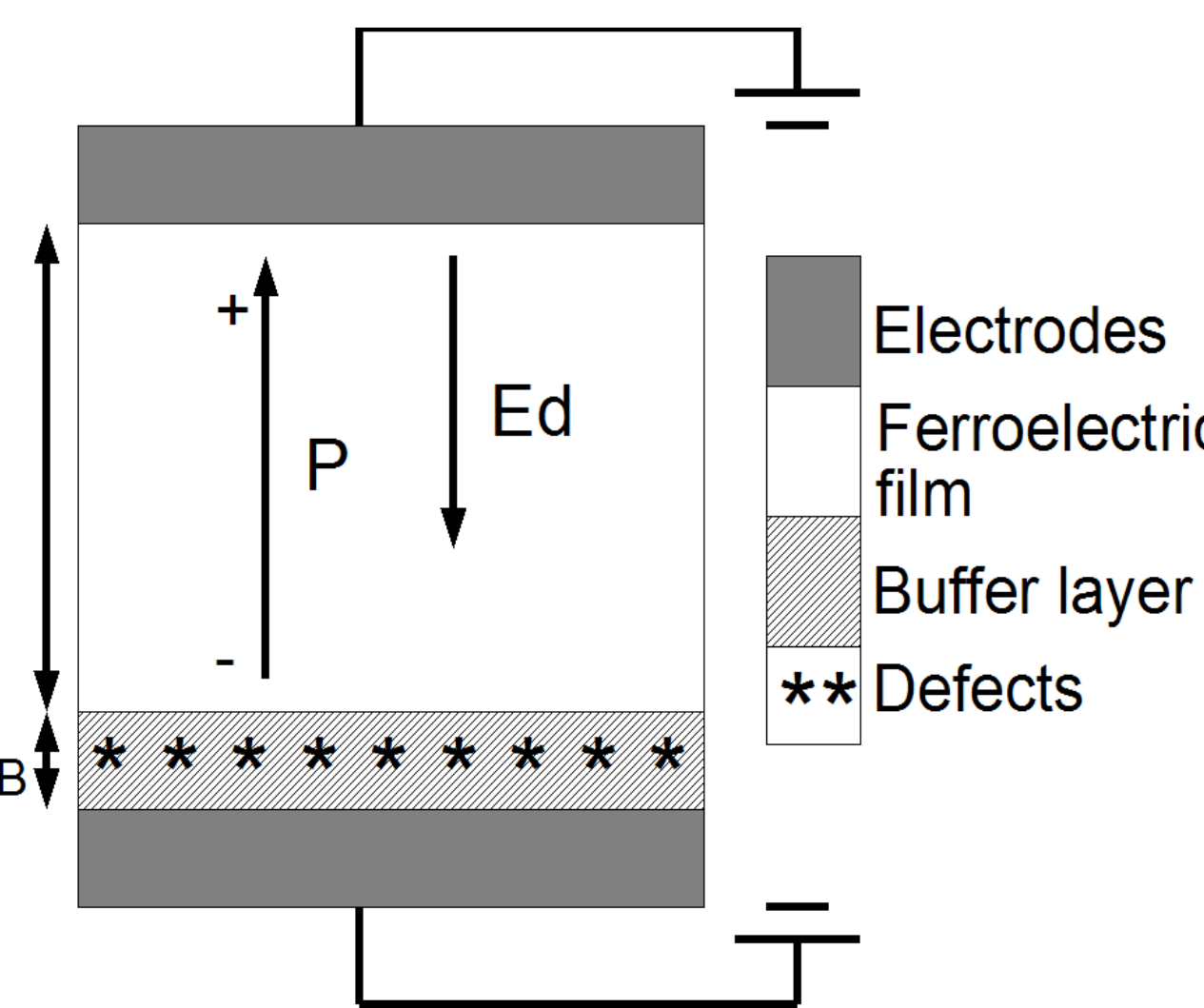
## Misfit dislocations

- Misfit strain from film-substrate interface:  $u_m = (b - a_0)/b$  (compressive  $u_m < 0$ , tensile strain  $u_m > 0$ )
- **Very thin films**: Uniformly strained by elastic deformation
- **Thick films**: Strain approaches zero far from substrate (bulk limit)
- Strain is gradually relaxed by forming dislocation lines
- **How are misfit dislocations distributed within the film (100-1000nm)?**
  - Uniformly: inhomogeneous strain
  - Segregated strain: dislocations reside within a thin buffer layer

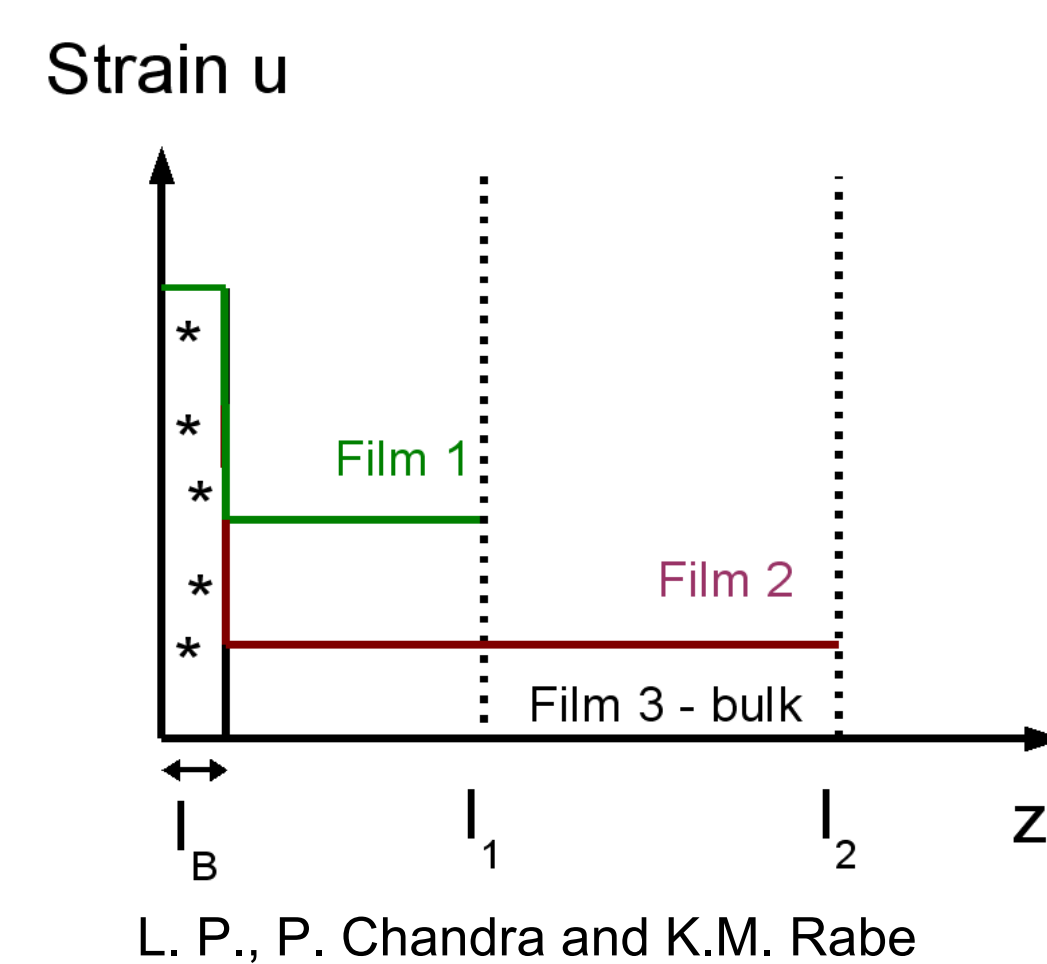


## Segregated strain model

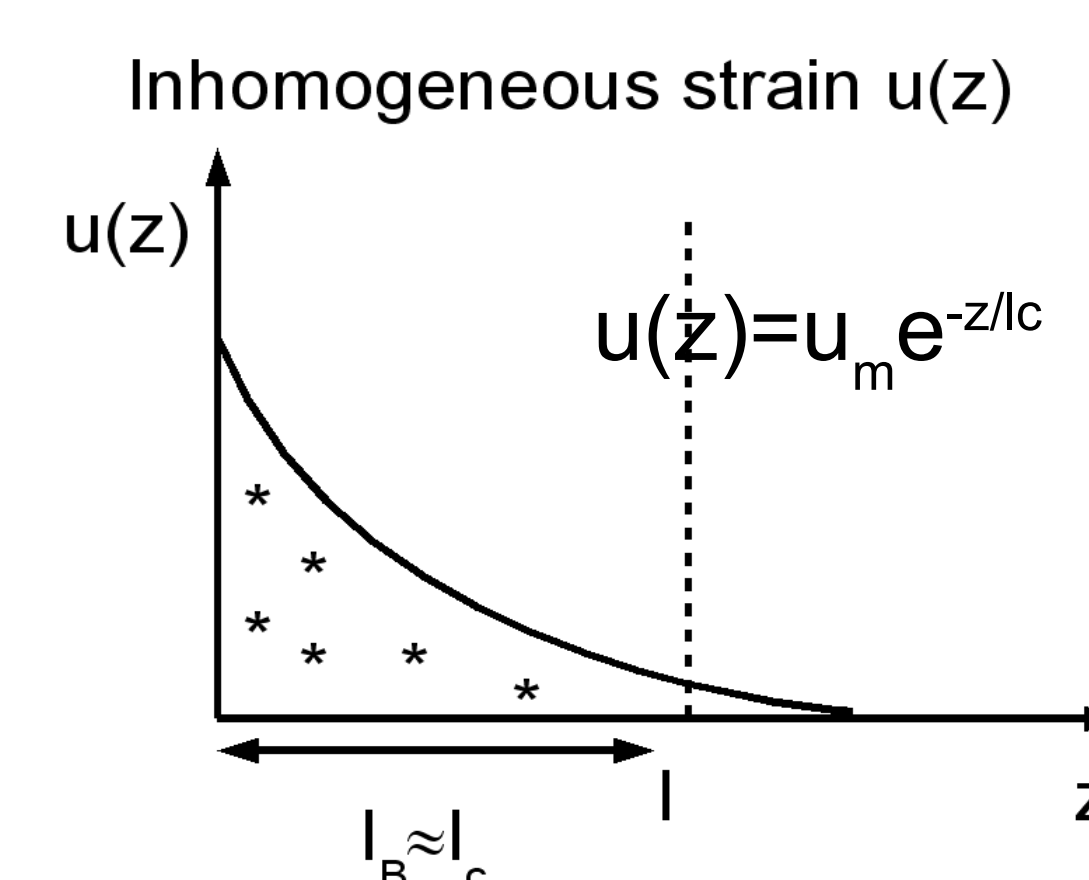
- Misfit dislocations reside within a thin buffer layer of the film,  $l_B \ll l$
- Elastic relaxation: homogeneous strain and polarization in the film (except buffer layer)
- More relaxation is observed for thicker films [model]:  $u_l = u_m e^{-z/l_c}, l_c \sim l$
- Planar ferroelectric single-domain film with uniaxial polarization
- Built-up of free surface charge at the ferroelectric boundaries results in a depolarization field,  $E_d \sim l_e/l$ ; (significant for  $l \sim 100$  nm)



## Homogeneous vs. inhomogeneous strain



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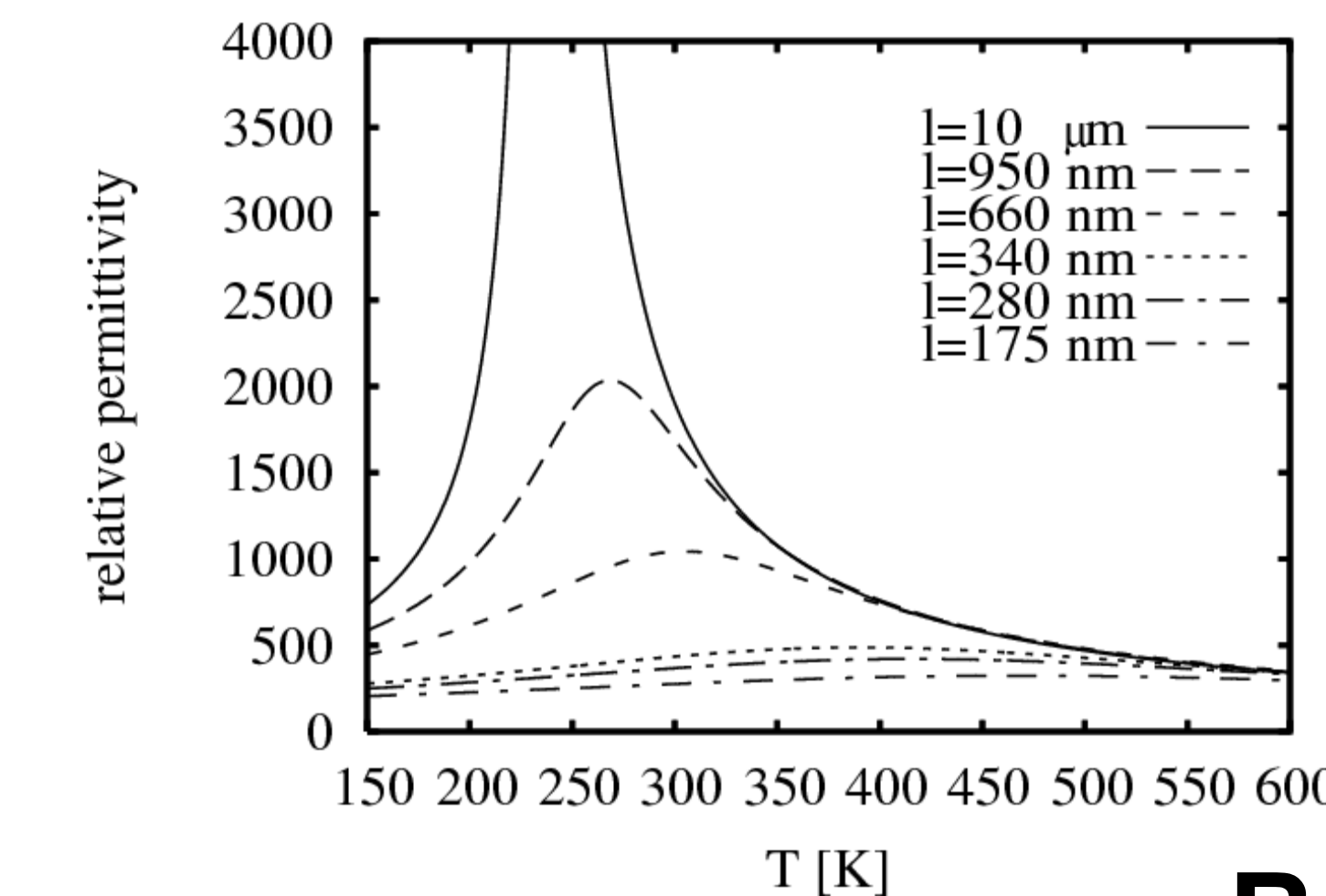
G. Catalan et al., J. Phys. Cond. Mat. **16**, 2253 (2004)

## Thickness dependent bias field ( $W_l$ )

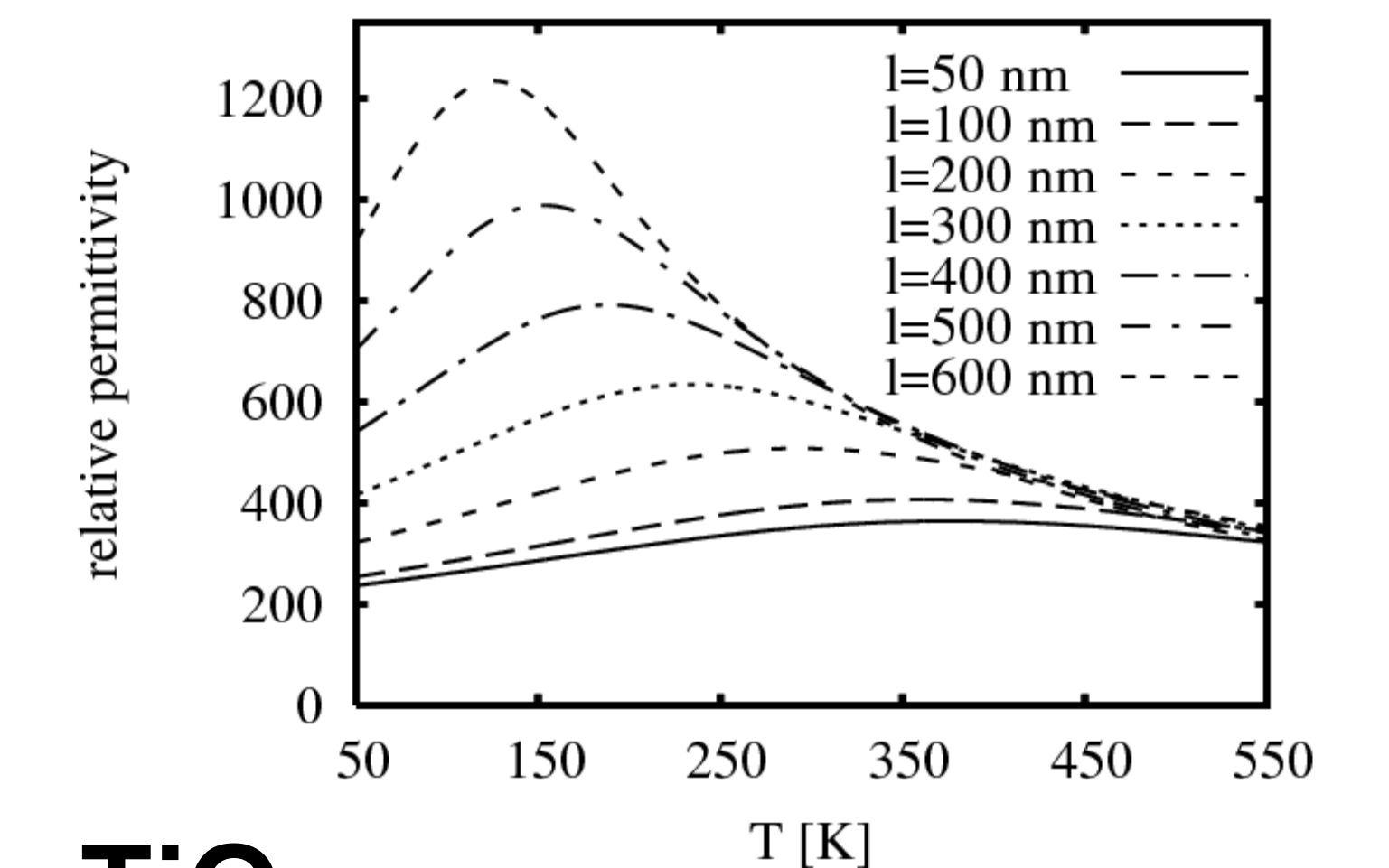
- We assume existence of a nonzero bias field ( $W_l$ ):
  - breaks  $P \rightarrow -P$  symmetry
  - broadens dielectric permittivity at phase transition
- We model thickness-dependence of  $W_l$  such that smearing of the permittivity is increased for thinner films:  $W_l = W_0 e^{-l/l_c}$
- We choose  $W_0 = 400$  kV/cm that is comparable to the amount of experimentally applied external electric field ( $E_{ext}$ )
- Bulk limit  $l \rightarrow \infty$ : permittivity diverges ( $2^{nd}$  order phase transition)

## Dielectric properties of $Ba_{0.5}Sr_{0.5}TiO_3$ (left) and strained $SrTiO_3$ (right)

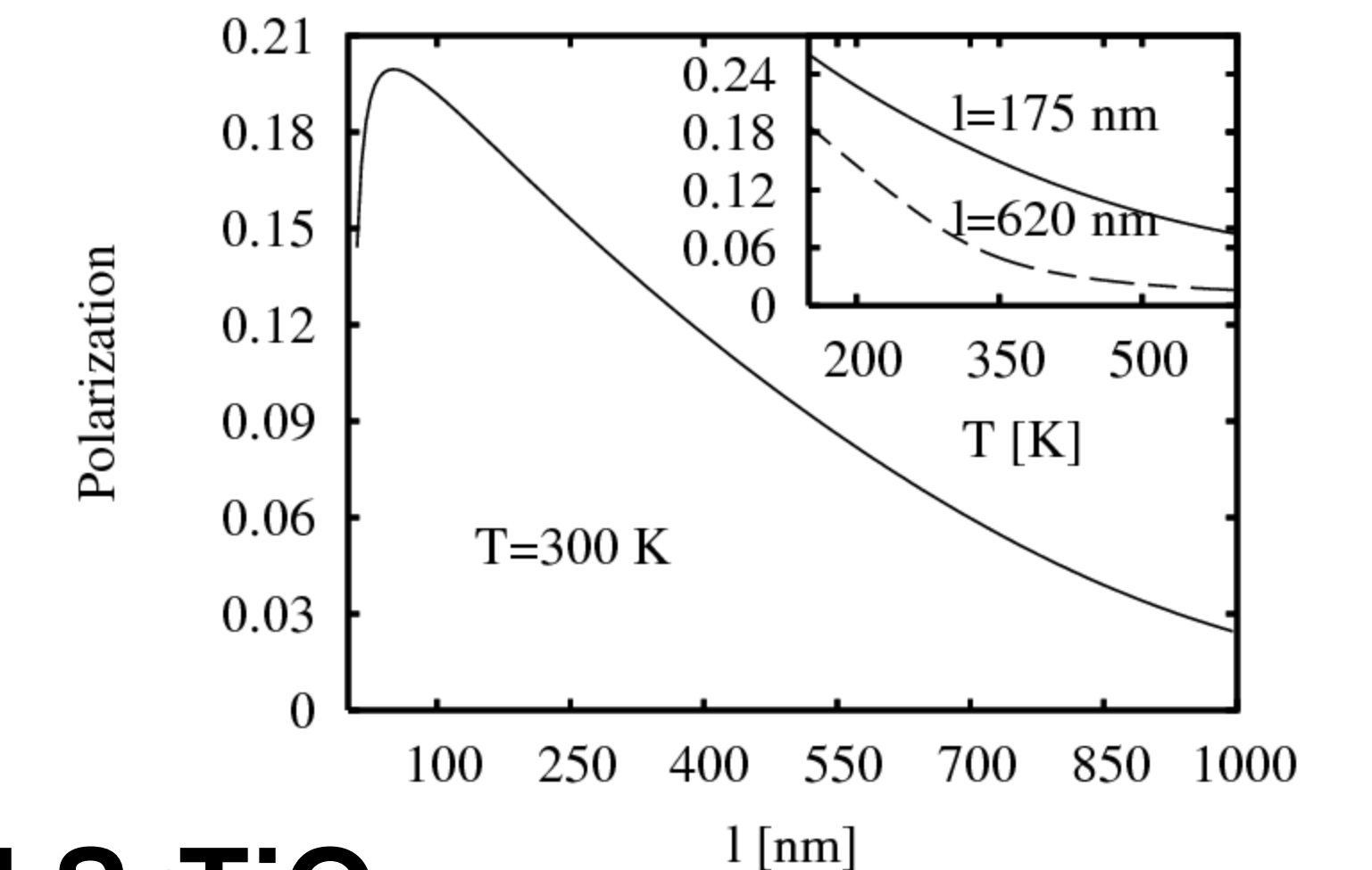
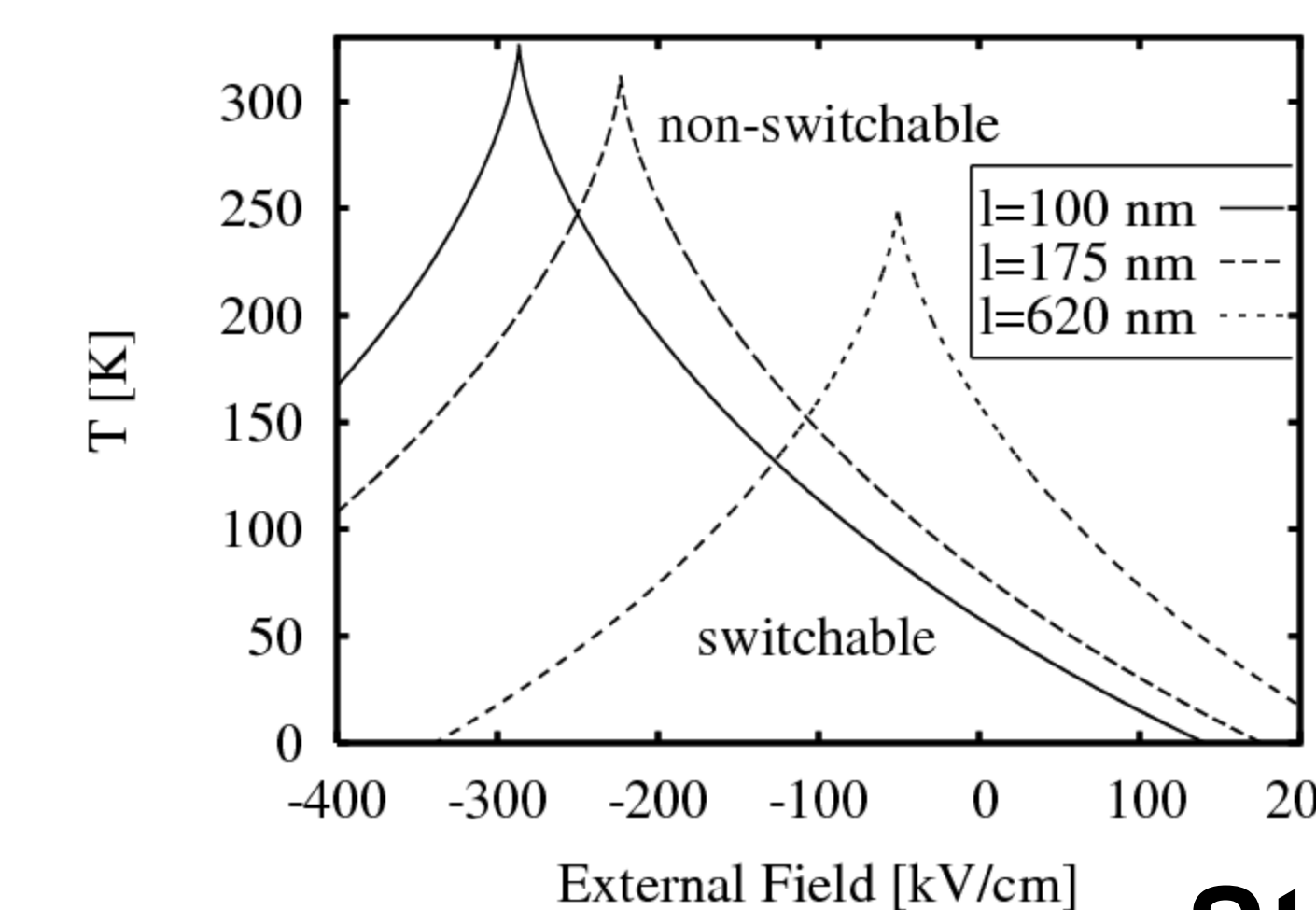
- Relative **permittivity** is **broadened** and more suppressed for thinner films
- Temperature at which the maximum of the permittivity occurs is shifted to higher values for thinner films in agreement with the experiment



$Ba_{0.5}Sr_{0.5}TiO_3$

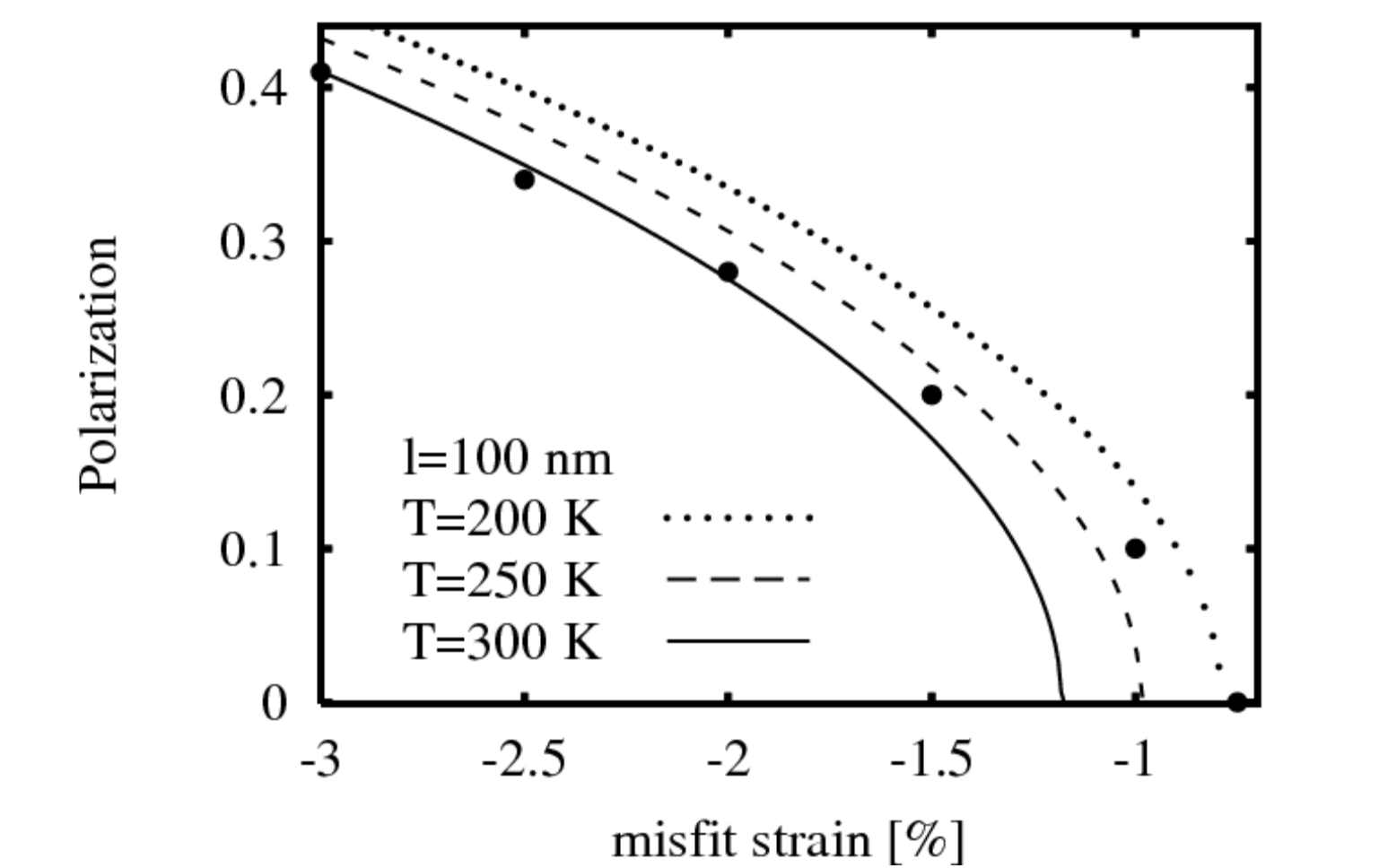
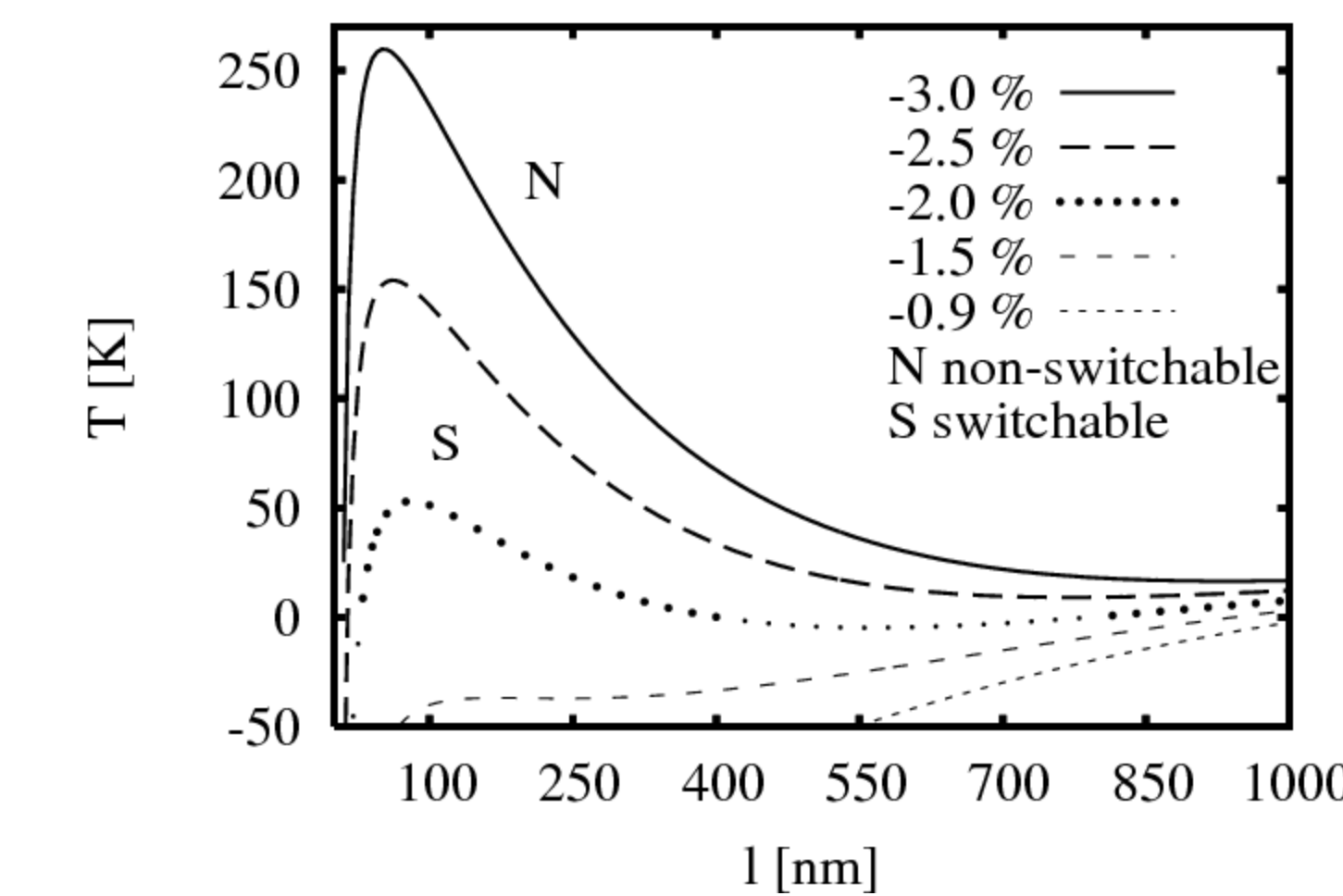


- $T_{onset}$  shows switchable polarization (hysteresis loops) at a given temperature in a certain region of the applied field ( $E_{ext}$ );  $W_l$  is field offset in the hysteresis loops
- Polarization as a function of film thickness and temperature **agrees with experiment**



**Strained  $SrTiO_3$**

- $T_{onset}$  as a function of film thickness **predicts ferroelectric phase for large enough strain**
- Polarization as a function of misfit strain is **compared to ab-initio calculations** (dots)



A. Antons et al., Phys. Rev. B **71**, 024102 (2005)

## Open questions

- How to experimentally distinguish between segregated model and inhomogeneous strain model?
  - Both models fit the experimentally observed dielectric properties
  - **Our prediction**: *observed sharpening* of the permittivity at zero 'net' field:  $E_{net} = 0 = E_{ext} + W_l$
- Origin of  $W_l$ : asymmetric boundaries due to buffer layer (edge dislocations);  $W_l \sim u_l$
- Curved boundary conditions: inhomogeneous strain is more pronounced