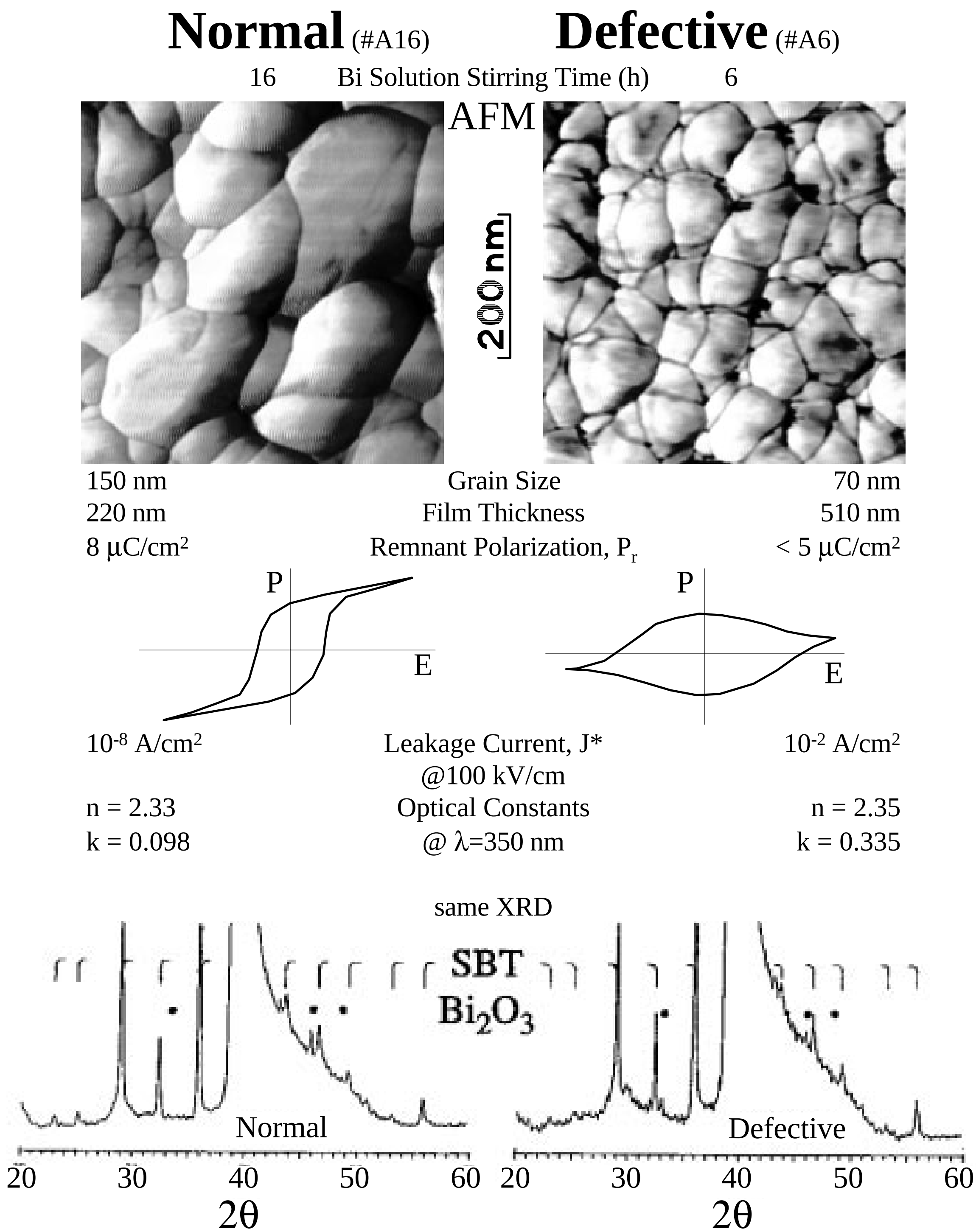


Figure 2-1. Comparing the properties of normal and defective $SrBi_2Ta_2O_9$ films with 13 molar % excess Bi. A normal SBT film has high Polarization, Pr, and low leakage current, J*, while a defective SBT film has high J*. XRD was not able to explain the different behavior of these two films which had the same stoichiometry, deposition method, and heat treatment. However, the defective film with 50 % smaller grains had a 340 % higher optical extinction coefficient.

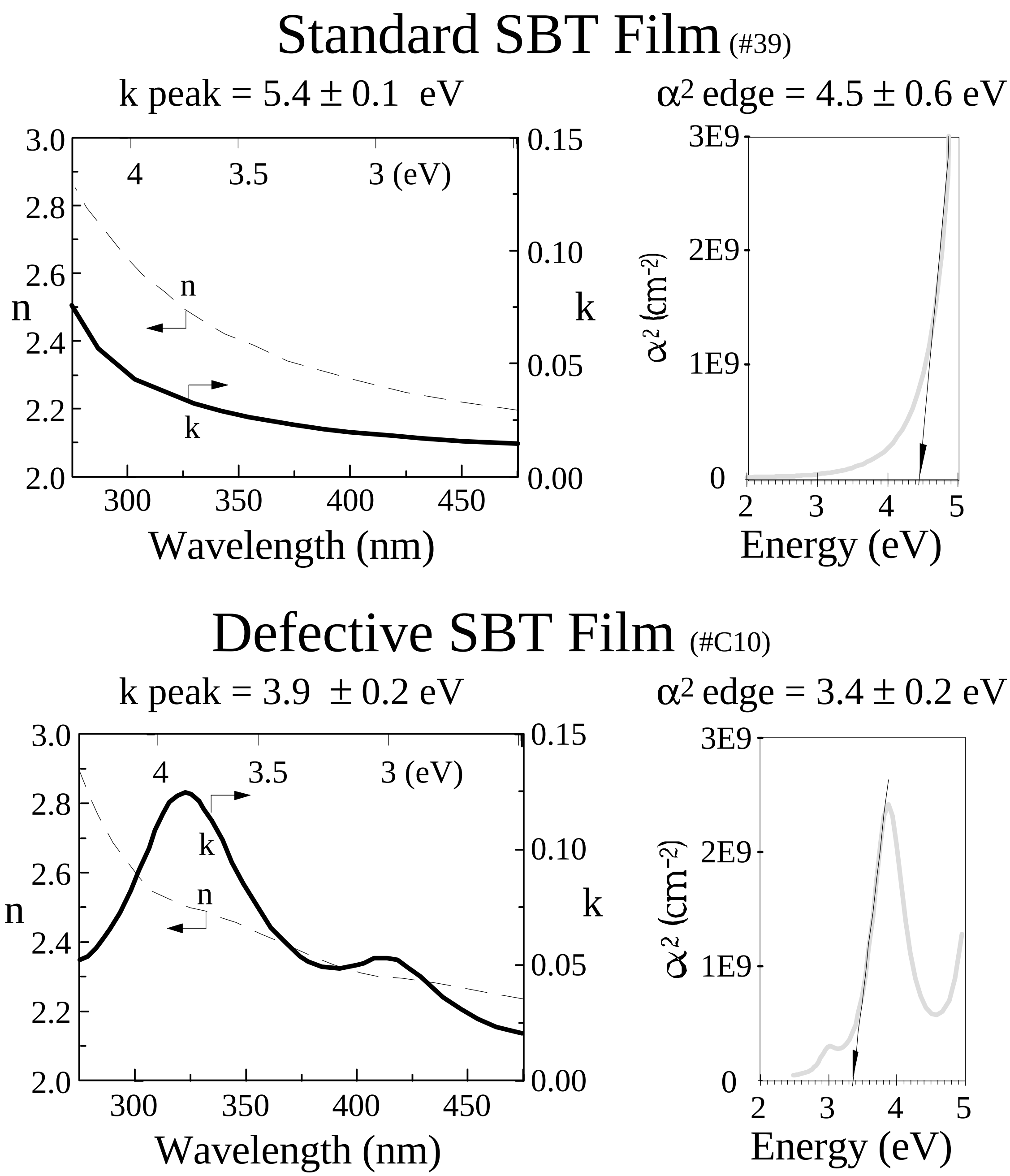


Figure 2-2. Optical dispersion of SBT films. A standard SBT film with excellent ferroelectric properties has a wide band gap absorption peak near 5.4±0.1 eV with an absorption edge at 4.5±0.6 eV. A defective SBT film also showed a strong absorption peak near 3.9±0.1 eV, with an absorption edge at 3.4±0.2 eV. Defective SBT films exhibited n and k spectra that were not characteristics of standard SBT; however, very similar features were found in Bi_2O_3.

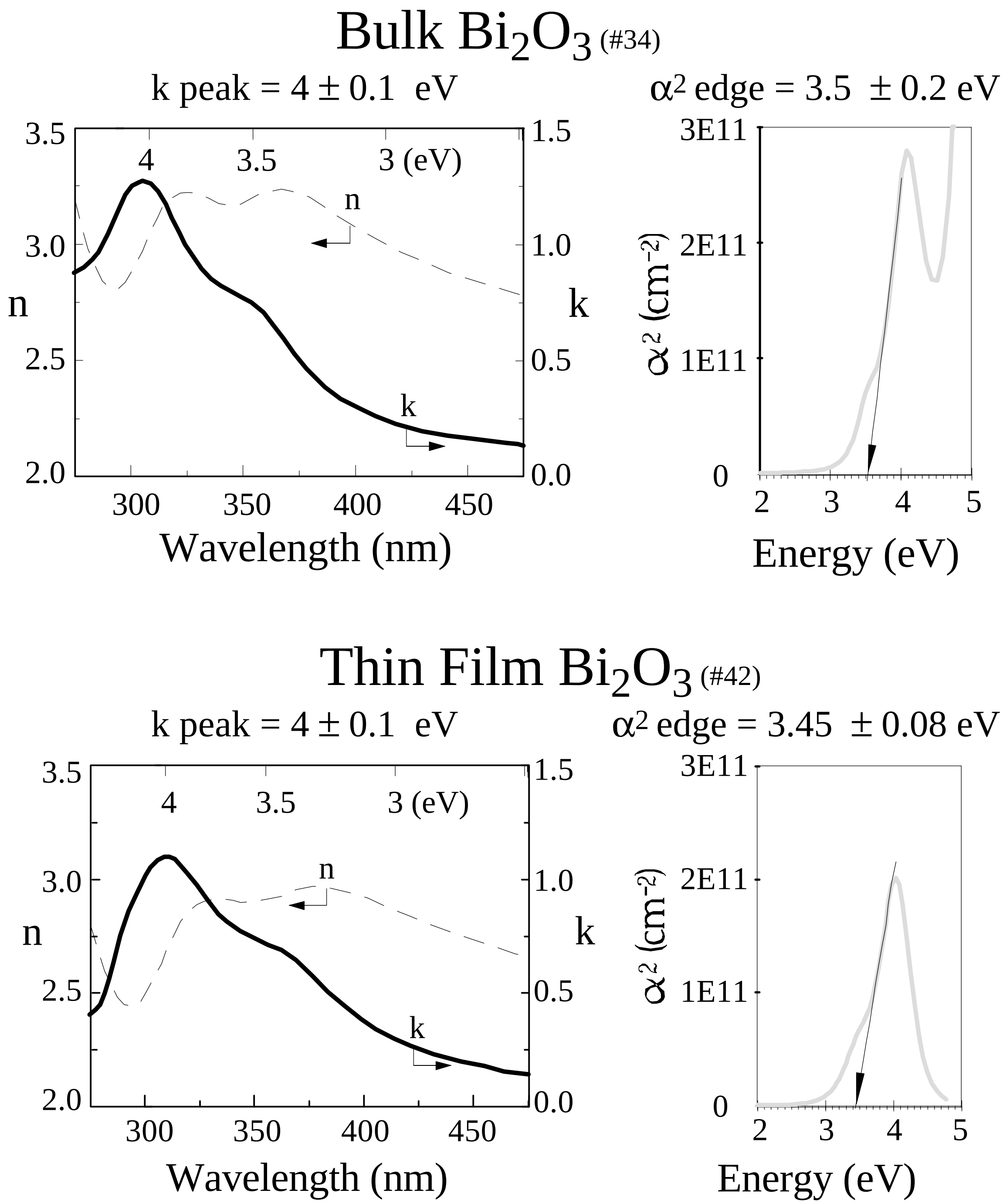


Figure 2-3. Optical dispersion of bulk (top) and thin film (bottom) Bi_2O_3. Both forms of Bi_2O_3 showed a strong absorption peak near 4.0±0.1 eV, with an absorption edge at 3.5± 0.2 eV for bulk and 3.45±0.08 eV for thin film Bi_2O_3. The fact that these features match the anomalous absorption in defective SBT films suggests that defective SBT films contain Bi_2O_3 as a separate phase.

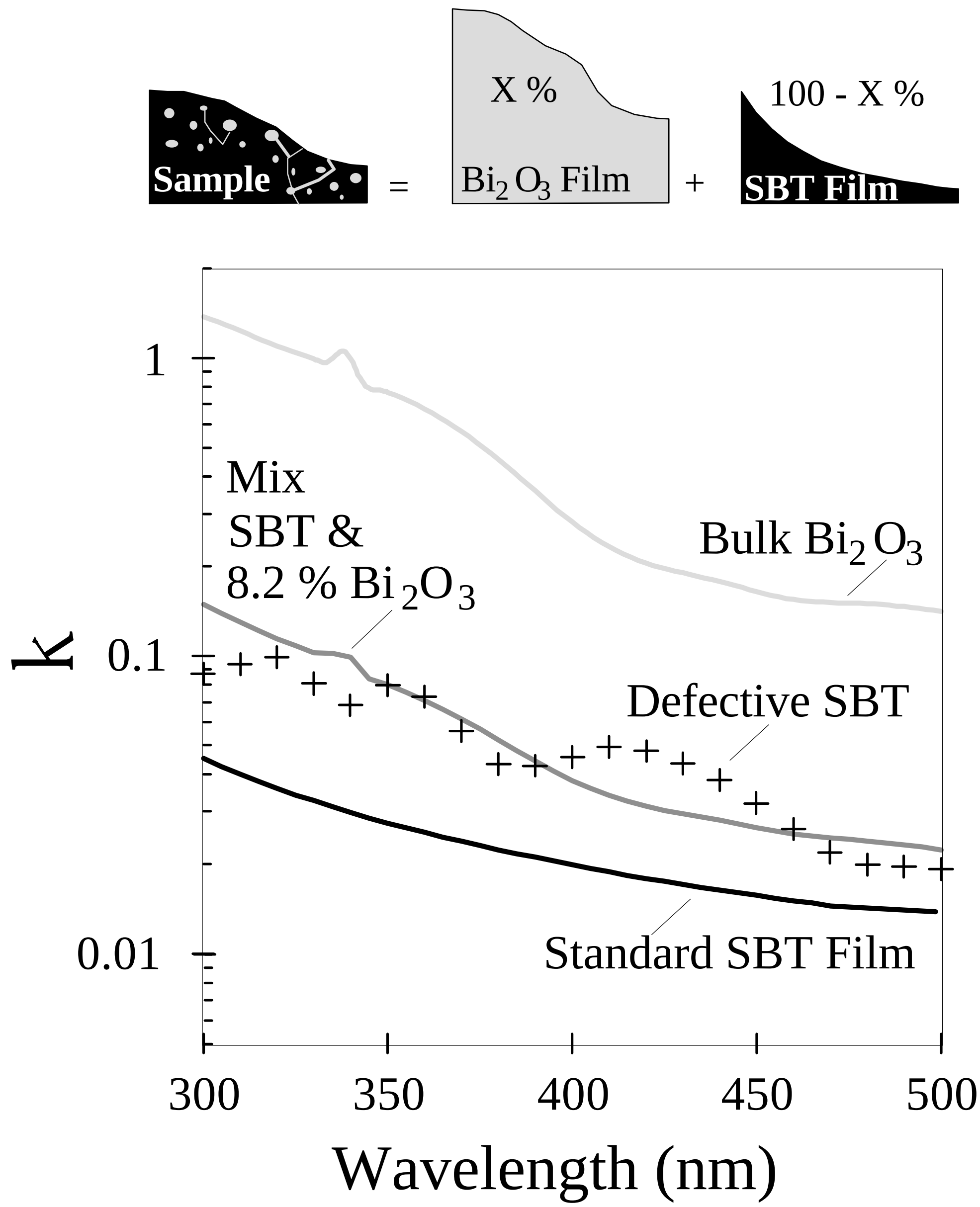


Figure 2-4. Estimating Bi_2O_3 content from optical dispersion. VASE spectra of thin film samples were modeled as Bruggeman EMA mixtures of X % bulk Bi_2O_3 and standard SBT film material. This method could detect small fractions of Bi_2O_3 because the extinction coefficient, k, of Bi_2O_3 was 10 times higher than that of standard SBT.

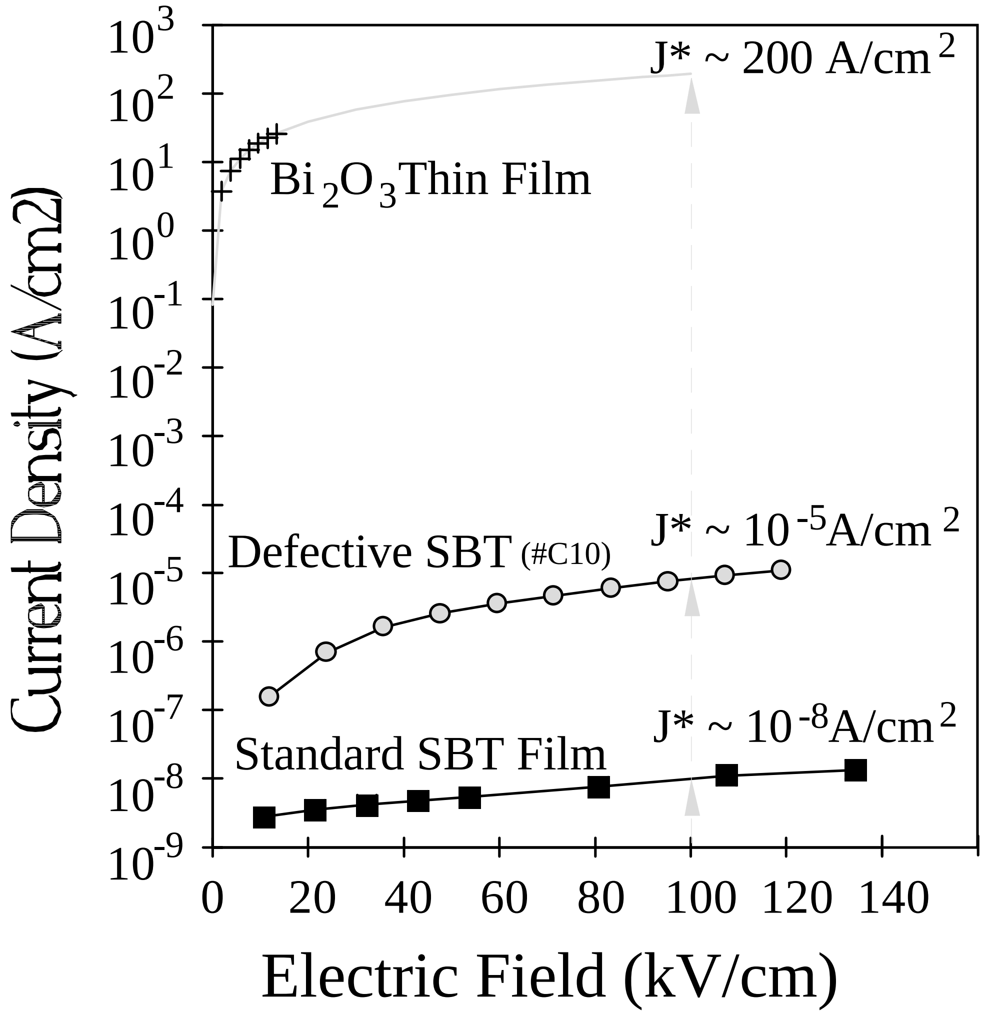


Figure 2-5. Leakage current of SBT and Bi_2O_3 films. Leakage current density at 100 kV/cm, J*, was estimated from the I-V characteristics of each sample. Since Bi_2O_3 has a high leakage current, the conductivity of a defective SBT film (#C10) with 8.2±1 % Bi_2O_3 was about 1000 times higher than a standard SBT film.

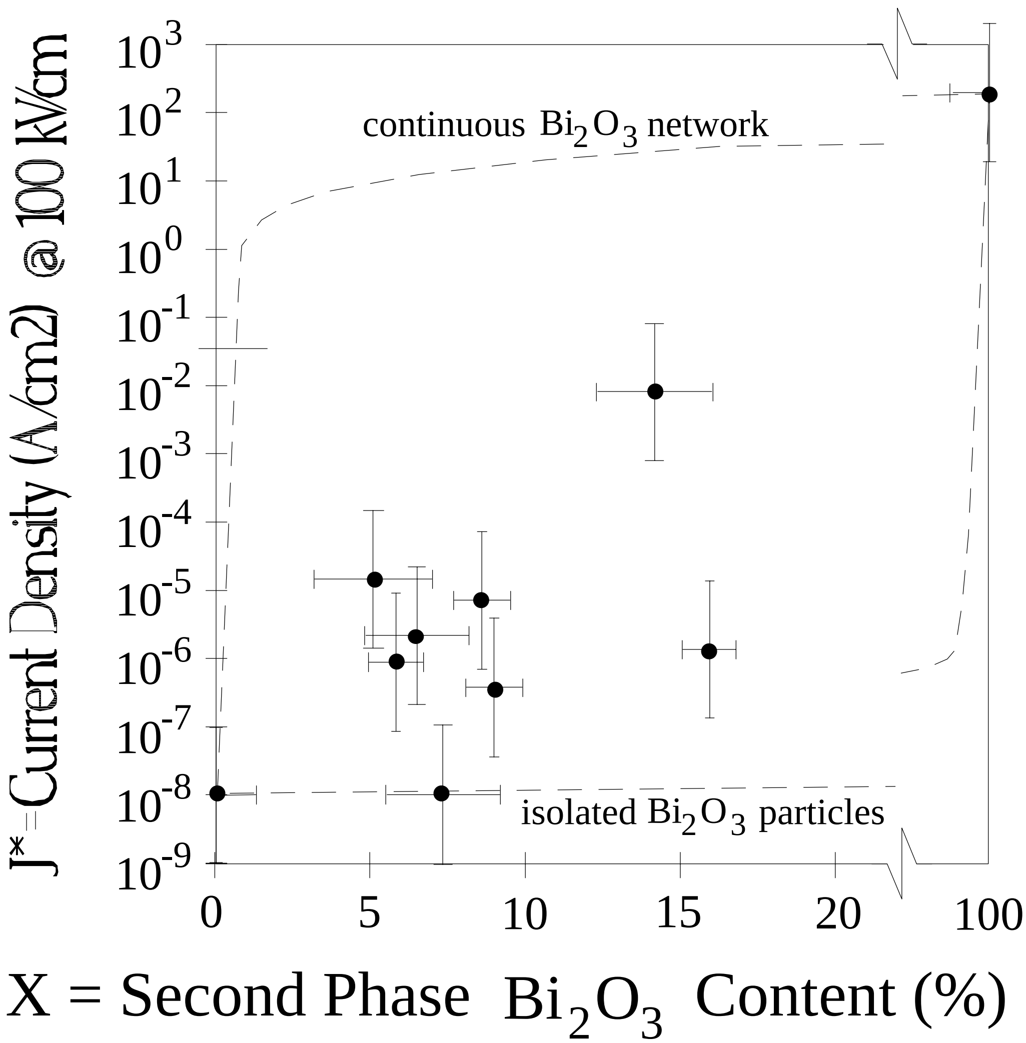
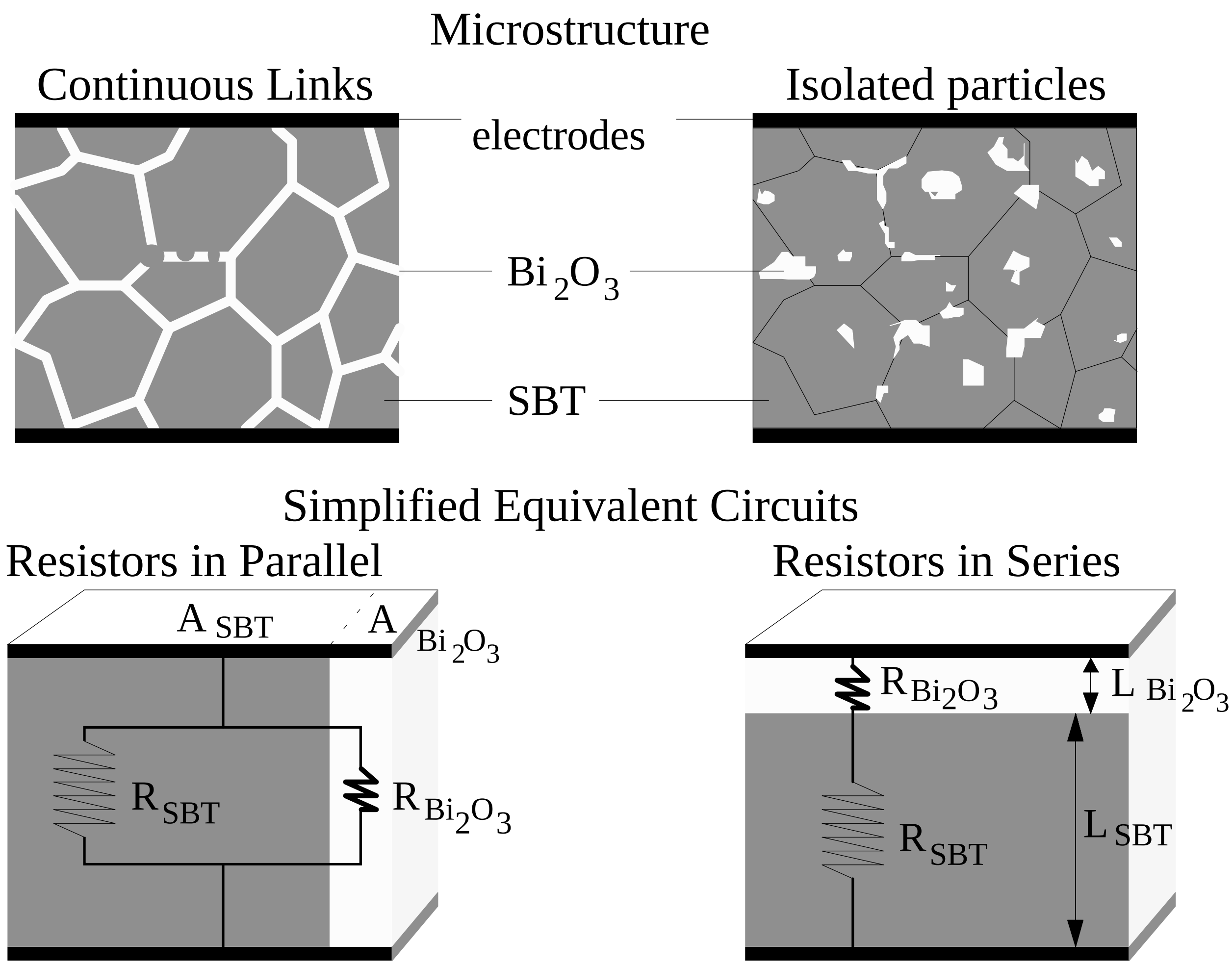


Figure 2-6. Optical explanation of conductivity. Films with 5% to 10% Bi_2O_3 (according to the VASE EMA method) had leakage currents up to 3 orders of magnitude higher than standard SBT, with J*~10^{-8} A/cm^2. In films with less than 10 % Bi_2O_3, the J* values are significantly lower than calculated for a continuous Bi_2O_3 network (at the grain boundaries, for example), indicating that Bi_2O_3 behaved more like isolated particles. Films made from solutions that were only stirred for 6 hours (total hours stirred is indicated by each data point) contained about 15 % Bi_2O_3 and had 2 to 6 orders of magnitude higher conductivity than standard SBT. The behavior of J* vs. X_{Bi2O3} is consistent with percolation models assuming Bi_2O_3 particles begin to form conductive links near threshold compositions, Xc, between 7 % and 18 % Bi_2O_3. The presence of optically detectable Bi_2O_3 generally explained high leakage currents in defective SBT films. However Bi_2O_3 content alone did not accurately predict leakage current. Other factors, such as particle geometry, seem to play a dominant role in determining conductivity.



Leakage Current Expressions

Upper Limit

$$I = I_{Bi_2O_3} + I_{SBT}$$

$$I \propto A_{Bi2O3} \propto X_{Bi2O3}$$

$$J^*_{net} = X_{Bi_2O_3}\, J_{Bi_2O_3} + X_{SBT}\, J_{SBT}$$

Lower Limit

$$R = R_{Bi_2O_3} + R_{SBT}$$

$$R \propto L_{SBT} \propto X_{SBT}$$

$$\frac{1}{J^*_{net}} = \frac{X_{Bi_2O_3}}{J_{Bi_2O_3}} + \frac{X_{SBT}}{J_{SBT}}$$

Figure 2-7. Effect of particle geometry on the net leakage current of composite films. If Bi_2O_3 forms a continuous network, then the maximum net conductivity is proportional to the conductive path's cross-sectional area, which scales linearly with X_{Bi2O3}. If Bi_2O_3 is in separated particles, then the maximum net resistivity is proportional to the length of the insulator's path, which scales linearly with the % of SBT, 100-X_{Bi2O3}. According to percolation theory, leakage current makes a transition from the lower limit to the upper limit as isolated particles merge into conductive links at some critical compositions.