

Studies of Dielectric Relaxation in  $\text{La}_{1-x}\text{Sr}_x\text{FeO}_3$  with  
Dielectric Time Domain Spectroscopy

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Dielectric time domain spectroscopy is a technique used in measurements of dielectric loss functions in a range from 1 MHz to 3 GHz [1]. Real time response  $V(t)$  of a sample to short rise time electrical potential ( $\sim 50$  psec) are recorded and Laplace transformed to the frequency domain,  $v(i\omega)$ . The complex permittivity or dielectric response  $\epsilon^*(i\omega)$  in the frequency domain for an open coaxial line sample cell design is

$$\epsilon_{\sigma}^* - \epsilon_r^* = \frac{c}{d} \frac{V_r - V_s}{i\omega(V_r + V_s)} z \cot z \quad (1)$$

where the subscripts r and s represent reference and sample, c is the speed of light in a vacuum, d the effective electric sample depth, and  $z = \frac{(\omega d)}{c} (\epsilon_r^* + \epsilon_s^*)^{1/2}$ . (The  $i\omega$ 's of the functions are left off). The usual reference is air,  $\epsilon_{\text{air}}^* = 1$ . If the dielectric loss function fits to

$$\epsilon^* = \epsilon_{\infty} + \frac{\epsilon_0 - \epsilon_{\infty}}{1 + i\omega\tau} = \epsilon_{\infty} + \frac{\epsilon_0 - \epsilon_{\infty}}{1 + (\omega\tau)^2} - i \frac{(\epsilon_0 - \epsilon_{\infty})\omega\tau}{1 + (\omega\tau)^2}$$

the sample exhibits Debye dielectric behavior, which is associated with the rotation of dipoles.

Application of electric field to a dielectric solid with partially mobile charges creates an added form of polarization to those normally associated with solid state properties. At a local level, the movement of an anion or cation to a previously vacant defect site can be pictured as dipole motion. The first report of this was by Breckenridge in 1948 as a study of alkali halides [2]. Since then there have been theoretical justifications for the assumption of Debye-like dielectric behavior in these systems [3,4,5].

The  $\text{La}_{1-x}\text{Sr}_x\text{FeO}_3$  systems are perovskites where the replacement of an  $\text{La}^{3+}$  with  $\text{Sr}^{2+}$  creates an  $\text{Fe}^{4+}$  for charge balance. Mössbauer studies [6] show the iron sites have an averaged oxidation state between +3 and +4 at room temperature because of thermal motion of the  $\text{Fe}^{4+}$  hole. Previous results on systems with  $0.1 \leq x \leq 0.3$  using TDS measured the thermal barriers to dielectric relaxation and conductivity and found  $\Delta E_D < \Delta E_{\text{con}}$ .

A signal averaging interface board attached to the time domain reflectometer (TDR) improved the accuracy and greatly speeded up the acquisition of results. Analysis of the spectra by equation 1 proved limited due to instrumental difficulties. The spectra are analyzed by simulation using a program designed to take Debye dielectric parameters,  $\epsilon_{\infty}$ ,  $\epsilon_0 - \epsilon_{\infty}$ , and  $\tau$ , produces  $V_s(t)$ . Results for  $\epsilon_{\infty}$

and any relaxation times less than 0.5 nsec are good, for  $0.5 \leq \tau \leq 1$  nsec they are marginal, and for longer  $\tau$  values, suspect at best.

Studies on systems with low dopant levels ( $x = 0.005, 0.01$ ) are done in order to determine the barriers to dielectric relaxation in systems where the defects are well separated. Both the 0.5% and 1% doped samples exhibit two distinct relaxation times at temperatures above 300K. The low frequency (long  $\tau$ ) relaxation has a larger loss ( $\epsilon_0 - \epsilon_\infty$  value) at temperatures between 300-340 in the 1% system but begins to drop at these temperatures. Analysis of the barrier gives a  $\tau_0$  value of 100 femtoseconds and a barrier of 0.22 eV. Estimates for the longer  $\tau$  relaxation gives  $\tau_0 = 50$  femtoseconds with the same barrier. In the 0.5% system the  $\tau_0$  and  $\Delta E$  values for the two relaxation times are 337 femtosec, 0.19 eV, 1.66 psec, 0.17 eV for the short and long frequencies, respectively.

No single reason is found for the behavior seen in these samples. Assigning a relaxation time to a certain phenomenon is difficult without more and better data. However, a mixture of the undoped LaFeO<sub>3</sub> with the doped systems show the low frequency relaxation mechanism is affected more than the high frequency mechanism. This may point to the long  $\tau$  being associated with surface polarization rather than bulk effects or to conductivity in the sample. Results on the effects of particle size and packing density proved inconclusive.

## References

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