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Electrical properties analysis of materials with ferroic order

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M. Smari et al have synthesized and investigated dielectric-electric properties of $\text{La}_{0.5}\text{Ca}_{0.5-x}\text{AgMnO}_3$ manganites [1]. In this letter we emphasize that the data analysis in the paper [1] is partially not reasonable.

First of all it should be admitted, that the Curie-Weiss law valid only for the static dielectric permittivity (i. e. $\varepsilon'(\omega \rightarrow 0)$ when $\varepsilon''/\varepsilon' \rightarrow 0$) and close to the phase transition temperature [2], while for the frequency dependent dielectric permittivity $\varepsilon'(\omega)$ should by more general approach applied, for example [3]:

$$\varepsilon^*(\nu) = \varepsilon_\infty + \Delta\varepsilon \int_{-\infty}^{\infty} \frac{f(\tau) d \ln \tau}{1 + (\omega\tau)^2}, \quad (1)$$

where $\varepsilon^*(\omega)$ is related with distributions of relaxation times $f(\tau)$. The electrical conductivity of $\text{La}_{0.5}\text{Ca}_{0.5-x}\text{AgMnO}_3$ is very high in all temperature range and at different frequencies, so that $\varepsilon''/\varepsilon'$ is also very high and no static dielectric permittivity data is presented in [1]. Moreover, far and very close to the phase transition temperature discrepancies from the Curie-Weiss law always appears even for pure ferroelectrics and these discrepancies are substantially temperature dependent [2]. Therefore, the conclusion in [1] that „in $\text{La}_{0.5}\text{Ca}_{0.1}\text{AgMnO}_3$ is observed the dielectric transition at 200 K, while such transition is not observed in $\text{La}_{0.5}\text{Ca}_{0.5}\text{AgMnO}_3$ “ is not reasonable.

Another interesting effect observed in $\text{La}_{0.5}\text{Ca}_{0.5-x}\text{AgMnO}_3$ is related with the maximum of the imaginary part of the complex dielectric permittivity ε'' , for $x=0$ its position is frequency dependent while for $x=0.4$ its position is frequency independent. In [1] it was wrongly explained by „relaxor dielectric behaviour“ for $x=0$ and „normal dielectric behaviour“ for $x=0.4$. It should be admitted that $\text{La}_{0.5}\text{Ca}_{0.5}\text{AgMnO}_3$ not demonstrate ferroelectric relaxors properties, because the loss tangent is very high and the maximum position of the real part of the complex dielectric permittivity is almost frequency independent. Why the dielectric behaviour of $\text{La}_{0.5}\text{Ca}_{0.5}\text{AgMnO}_3$ and $\text{La}_{0.5}\text{Ca}_{0.1}\text{AgMnO}_3$ is different? The complex dielectric permittivity $\varepsilon^* = \varepsilon' - i\varepsilon''$ is related with the complex electrical impedance $Z^* = Z' - iZ''$:

$$\varepsilon' = \frac{Z''}{\varepsilon_0 \omega ((Z')^2 + (Z'')^2)}, \quad (2)$$

$$\varepsilon'' = \frac{Z'}{\varepsilon_0 \omega ((Z')^2 + (Z'')^2)}. \quad (3)$$

Often for conductors when $\omega \rightarrow 0$, Z' reach its static value Z'_{stat} related with the dc conductivity and $Z'' \approx 0$. In this case $\varepsilon'' = 1/(\varepsilon_0 \omega Z') = \sigma_{\text{dc}}/\varepsilon_0 \omega$ (where σ_{dc} is the dc conductivity) and the temperature behaviour of dielectric losses ε'' resembles the temperature behaviour of the dc conductivity. This situation is clearly observed for $\text{La}_{0.5}\text{Ca}_{0.1}\text{AgMnO}_3$. However, for $\text{La}_{0.5}\text{Ca}_{0.5}\text{AgMnO}_3$ Z'' is not zero even at low frequencies (below 100 Hz in [1]) in this case the temperature behaviour of the

dielectric losses ϵ'' is substantially frequency dependent. For example, for resistivity (R) and capacitance (C) connected in serial the complex electrical impedance is:

$$Z' = \frac{Z_{stat}}{1+(\omega\tau)^2}, \quad (4)$$

$$Z'' = \frac{Z_{stat}\omega\tau}{1+(\omega\tau)^2}. \quad (5)$$

By combination Eqs. (2-5) it possible to conclude, that in this case the maximum position of dielectric losses ϵ'' should be frequency dependent. Therefore, the difference in the electric behaviour between $\text{La}_{0.5}\text{Ca}_{0.1}\text{AgMnO}_3$ and $\text{La}_{0.5}\text{Ca}_{0.5}\text{AgMnO}_3$ are mainly related with different values of the dc conductivity and the critical frequency (the frequency at which the value of the conductivity $\sigma(x)$ deviates from the dc plateau). Moreover, below room temperature the antiferromagnetic phase transition was observed in $\text{La}_{0.5}\text{Ca}_{0.5-x}\text{AgMnO}_3$ [4]. The anomalies of the conductivity and the dielectric permittivity (the last is related with the conductivity via the Kramers-Kronig relation) observed in $\text{La}_{0.5}\text{Ca}_{0.5-x}\text{AgMnO}_3$ are related with this antiferromagnetic phase transition. Can it be observed of the multiferroic behaviour in $\text{La}_{0.5}\text{Ca}_{0.5-x}\text{AgMnO}_3$? In order to answer to this question measurements at higher frequencies (where $\epsilon''/\epsilon' \rightarrow 0$) should be performed. This research is funded by the European Social Fund under the Global Grant measure.

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