POLARIZATION DYNAMICS IN $La_{5/3}Sr_{1/3}NiO_4$

N. Hakim*, Z. Zhai*, C. Kusko*, P.V.Parimi*, S-W. Cheong**, and S. Sridhar*†.

*Physics Department, Northeastern University, 360 Huntington Avenue,Boston,MA 02115

**Department of Physics and Astronomy, Rutgers University, Piscataway, NJ 08855.

ABSTRACT

Dynamic susceptibility measurements at microwave frequencies ($2-10GHz$) are a sensitive probe of charge dynamcis in $La_{5/3}Sr_{1/3}NiO_4$. Below the charge ordering temperature of $240K$, a dielectric loss peak due to a relaxation mode with a large dielectric susceptibility is observed, and is associated with charge stripe formation. The dielectric response for $H_\omega || b$ ($E_\omega \perp b$) is well represented by $\tilde{\varepsilon}(T) = \varepsilon_o/(1 - i\omega\tau(T))$, with $\varepsilon_o \sim 50$, and $\tau(T) = 2 \times 10^{-9}(\mathrm{sec}) \exp(-T/37K)$. Parallel conductivity $\sigma(T)$ contributions dominate at higher temperatures and for $H_\omega || c$ ($E_w \perp c$). The dielectric loss peak observed indicates that the charge relaxation rates lie in the GHz frequency ranges.

INTRODUCTION

The carrier doping in Mott insulators such as $La_2MO_{4+\delta}$ ($M = Ni, Cu$) has attracted much attention bacause of the existence of the high T_c superconductivity in hole doped La_2CuO_4. These quasi-two dimensional electronic systems exhibit charge and spin correlations in which doped holes tend to undergo stripe like ordering on the domain walls of antiferromagnetic stripes[1, 2, 6]. The evidence of dynamic stripes (spin fluctuations) in superconducting $La_{2-x}Sr_xCuO_{4-\delta}$ and static stripe correlations in nonsuperconducting $La_{2-x}Sr_xNiO_4$ has generated compelling interest about the interplay of stripe dynamics and superconductivity.

A variety of measurements have revealed three successive transitions associated with quasi-two dimensional commensurate charge ordering (at $\sim 240K$) and spin stripe ordering ($\sim 190 - 160K$) [1, 10, 9, 8, 2] in $La_{5/3}Sr_{1/3}NiO_4$. The spin ordering at $190K$ is driven by charge ordering when the ordering between charge stripes takes place. The existence of stripe glass is proposed in the temperature regime, $240-190K$[2, 9, 8, 11], but the details of orientational order is missing probably due to the dominant effect of short stripes. A key issue that has arisen is the role of the measurement time-scale since stripes, and more generally magnetic and charge correlations, are now believed to have strong dynamical properties, and previous measurements have been principally carried out with probes having very different time scales such as Neutron scattering ($\sim 10^{-13}$ sec) and NMR ($\sim 10^{-7}$ sec).

In this paper, we present measurements of the dynamic (microwave) response of $La_{5/3}Sr_{1/3}NiO_4$ using a precision superconducting microwave cavity at $10GHz$, supplemented by measurements at $2GHz$ using a normal Cu resonator. Our measurements probe short time ($\sim 10^{-11}$ sec) or high frequency dynamics of charge correlations in this material. We find that charge ordering at $\sim 240K$ suppresses the dia-electromagnetic contribution caused by eddy currents due to the conductivity. Instead, charge ordering is accompanied by the onset of a dynamic dielectric susceptibility, which freezes out (quasi-statically at the finite measuring frequency) as the temperature is lowered due to rapid increase of the relaxation time. Our results can be succinctly summarized in terms of a T-dependent conductivity $\sigma(T)$ and a dielectric constant $\tilde{\varepsilon}(T) = \varepsilon_o/(1-i\omega\tau(T))$. The charge relaxation time $\tau(T)$ increases exponentially with decreasing temperature T. A quantitative fit to the data is obtained with the form $\tau(T) = \tau_o \exp(-T/T_o)$.

The $La_{5/3}Sr_{1/3}NiO_4$ single crystals were prepared using a floating zone technique. Details of the crystal growth are given elsewhere[2]. The high quality of these crystals is well established by thorough characterization by several techniques[2].

EXPERIMENT

The principal measurements reported here are carried out using a superconducting microwave cavity. The superconducting cavity is made of Niobium,which is a superconductor below $T_c = 8.9K$. The dimension of the cylindrical cavity are: radius R=2.22 cm, and a length of L = 2.54cm. The TE_{011} mode resonates at $10GHz$. The sample supported on a sapphire rod is inserted and centered through a hole made at the bottom of the cavity. The entire cavity and assembly is vacuum tight, and in turn is placed in a bath of liquid $_4He$. To heat up the sample to higher temperature, the sapphire rod is thermally isolated from the cavity walls and a 50Ω heating coil is used to control the sample temperature from $4K - 300K$. The high quality factor $Q \sim 2 \times 10^8$ enables us to perform high precision microwave measurements. These experiments have been extensively utilized previously for measuring a variety of materials, including superconducting cuprate, manganate and borocarbide crystals [3, 4]. In all of the measurements, the sample (typically $2 \times 2 \times 1\ mm^3$) was placed at the center of the cavity where the H_ω is maximun and $\mathrm{E}_\omega = 0$ fot the TE_{011} mode.

The copper split ring resonator has a cylindrical shape with a split along the side. The inner radius $R_{in} = 0.395cm$, the outer radius $R_{out} = 1.037cm$, and the split gap is of thickness $= 0.07cm$. These dimensions give a resonance frequency of $2GHz$, and the quality factor $Q \sim 2000$. The resonator is placed within a conducting cylinder to maintain highest Q factor. Heating and supporting the sample inside the ring is done in a way similar to that supperconducting cavity discussed above.

We define an electromagnetic susceptibility $\tilde{\zeta}_H$which is obtained from the measured cavity resonance parameters by : $f(0) - f(T) + i\Delta f(T) = g[\zeta'_H(T) + i\zeta''_H(T)]$. Here $f(T)$ is the resonant frequency, $\Delta f(T)$ is the width of the resonance, and g is a sample geometric factor, assuming the sample is in the shape of a sphere. When the sample is placed in the center of the cavity and the TE_{011} mode is used, so that the sample is at a region of maximum microwave magnetic field, careful analysis of the cavity perturbation equations shows that the measured EM susceptibility $\tilde{\zeta}_H$ (we use the subscript H to denote that the sample is placed in an H field region), is related to the magnetic ($\tilde{\chi}_M$) and dielectric ($\tilde{\chi}_P$) susceptibilities in the following ways [5]:

$$\begin{aligned}
\tilde{\zeta}_H &= \tilde{\chi}_M && (k_o a)^2 \tilde{\chi}_E << \chi_M && (1)\\
&= \frac{1}{10}(k_o a)^2 \tilde{\chi}_P && (k_o a)^2 \tilde{\chi}_P >> \chi_M && (2)\\
&= \frac{3}{2}\left(1 - \frac{3}{(ka)^2} + \frac{3\cot ka}{ka}\right) && \text{arbitrary } ka && (3)
\end{aligned}$$

where $k^2 = k_o^2 \left(\tilde{\varepsilon} + i \frac{\sigma}{\omega \varepsilon_o} \right)$, $\tilde{\varepsilon} = \varepsilon' + i\varepsilon'' = 1 + \chi_P' + i\chi_P''$ is the complex dielectric constant, a is the sample diameter. Note that $\tilde{\zeta}_H$ represents the effective susceptibility which can include eddy current (or conductivity) contributions in addition to dynamic dielectric and magnetic response. The experiment measures the magnetic susceptibility $\tilde{\chi}_M$ only if the dielectric and conductivity contributions are negligible. Since the sample size is typically $2mm$, and hence $k_0 a \sim 0.2$, the dielectric contribution dominates if $\chi_P/\chi_M > 250$. This condition appears to be met in most of the oxides which are even slightly doped and/or weakly conducting, and certainly at high temperature. Thus although the sample is placed in a magnetic field region, at these high frequencies we mostly measure the dielectric (polarization) and/or conductivity dynamics rather than the spin dynamics. Only in very high resistance insulators, such as Sr_2CuO_3 and $ZnCr_2O_4$, and possibly at very low $T < 20K$ in $La_{5/3}Sr_{1/3}NiO_4$, we are possibly measuring the magnetic susceptibility at these high frequencies. The results for $\tilde{\zeta}_H$ can be used to extract information regarding the dielectric permittivity $\tilde{\varepsilon}$ and the conductivity σ.

While the loss term $\zeta''(T)$ is measured absolutely, the technique yields changes $\delta\zeta_H'(T) = \zeta_H'(T) - \zeta_H'(3K)$ in susceptibility with very high precision. In the present measurements since $\zeta_H'(T \to 0) \to 0$, $\delta\zeta_H'(T) \sim \zeta_H'(T)$ does represent an absolute measure of $\zeta_H'(T)$ for most of the temperature range in this work. Comparison of absolute values of the present microwave susceptibility with dc magnetic susceptibility $\chi_M(f = 0, T)$ reveals that we are observing completely new phenomena at these frequencies. Our experiments also enable us to measure the anisotropy by varying the microwave magnetic field direction (H_ω) with respect to the crystal axes $(a, b$ or $c)$.

RESULTS

The $10GHz$ susceptibility, $\zeta_{H,b}''(10GHz, T)$ for $H_\omega \,//\, b$ is shown in Fig. 1(a). A sharp drop is seen from $300K$ which is arrested around $240K$ and followed by a peak in the absorption at around $210K$. Further decrease in T results in monotonic decrease of $\zeta_{H,b}''$. The features observed in $\zeta_{H,b}''$ are reflected in $\delta\zeta_{H,b}'$. As can be seen from Fig. 1(b), the high temperature conductivity response which is dia-electromagnetic, is dominant above $240K$, and at lower T the dielectric response takes over, resulting in a peak at $240K$.

We have also carried out measurements at $2GHz$ to investigate the frequency dependence and anisotropy of the microwave features. Overall the features observed at $10GHz$ are reproduced at this frequency as seen in Fig. 2, in the $\zeta_{H,b}''(2GHz, T), H_w//b$ data. However, the absorption peak which appeared in the $10GHz$ data around $210K$ has moved down to $150K$, indicating a clear frequency dependance to this process.

A quantitative fit of the data to Eq(3) is obtained using two contributions which can be represented as $\sigma(T) - i\omega\tilde{\varepsilon}(\omega, T)$, which are:

1. a complex T-dependent dielectric function $\tilde{\varepsilon} = \varepsilon' + i\varepsilon'' = \varepsilon_o/(1 - i\omega\tau(T))$. Best fits to the data below $240K$ give $\varepsilon_o = 50$, $\tau(T) = \tau_o \exp(-T/T_{\tau o})$, with $\tau_o = 2 \times 10^{-9}$ sec, and $T_{\tau o} = 40K$.
2. a T dependent conductivity $\sigma(T) = \sigma_o \exp(-T_{\sigma o}/T)$, with $\sigma_o = 5 \times 10^6 (\Omega - m)^{-1}$ and $T_{\sigma o} = 3160K$. This conductivity value is intermidiate between the measured dc conductivity along the c-axis and the a-b (Ni-O) plane.

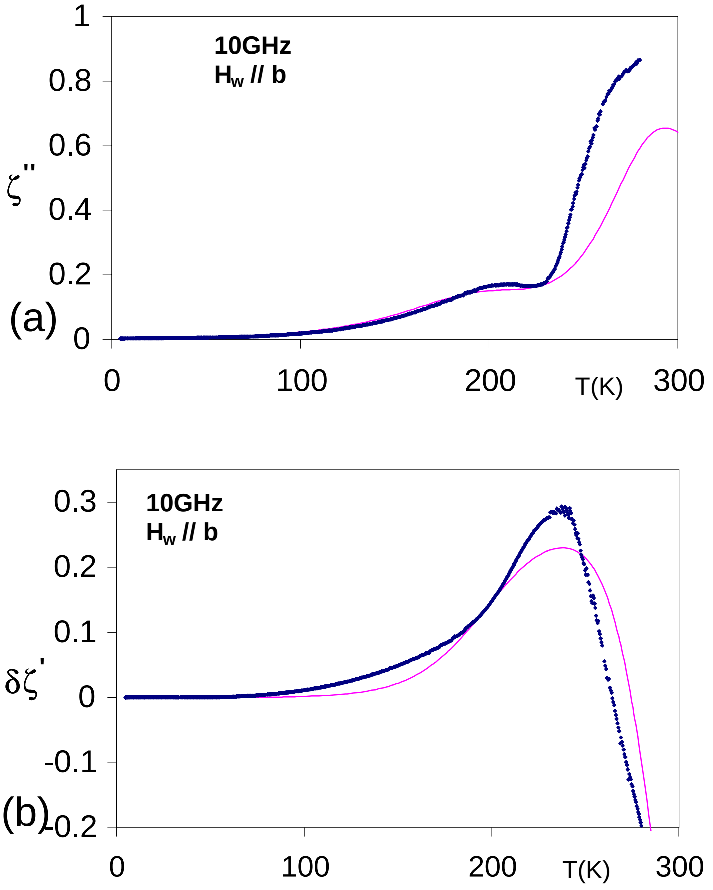

Figure 1: Microwave susceptibility $\zeta''_{Hb}(10GHz,T)(a)$ and $\delta\zeta'_{Hb}(10GHz,(T)(b)$ versus T for $La5/3Sr1/3NiO4$. Solid lines represent he susceptibility calculated from the dielectric and eddy current conributions. The dielectric loss peak at around 210K is visible in $\zeta''_{Hb}(10GHz,T)$ data and the model calculations.

The dielectric loss peak occurs at a peak temperature T_p where $\omega\tau(T_p) = 1$. For our data, $T_p = 210K$ at $10GHz$, and $T_p = 150K$ for the $2GHz$. The comparison of this model using eq.1 and including the above conductivity and dielectric contributions is shown in Fig 1 &2, and is seen to describe all the essential features of the data.

When $H_\omega \,||\, c$ at $2GHz$, Fig. 3a shows an absorption peak around $230K$ in $\zeta''_{H,c}(2GHz,T)$. This feature is reflected as a decrease in $\zeta'_{H,c}$ as a change of state to dia-electromagnetism. Knowing that the conductivity [12] is larger along the a-b plane, the nickel-oxygen plane, indicates that this response is mainly due to in-plane conductivity contributions. There is a small dielectric contribution also in this configuration at lower T. In the $H_w//b, 10GHz$ & $2GHz$, the dielectric contribution below $230K$ is more clearly visible since the conductivity is less along the c-axis. Above $230K$, where the charges are free, eddy current response dominates which explains the increase in absorption at high temperature.

A clear theme is emerging from the present measurements on $La_{5/3}Sr_{1/3}NiO_4$ when viewed with an extensive set of data taken by us on other oxides[7], including the spin

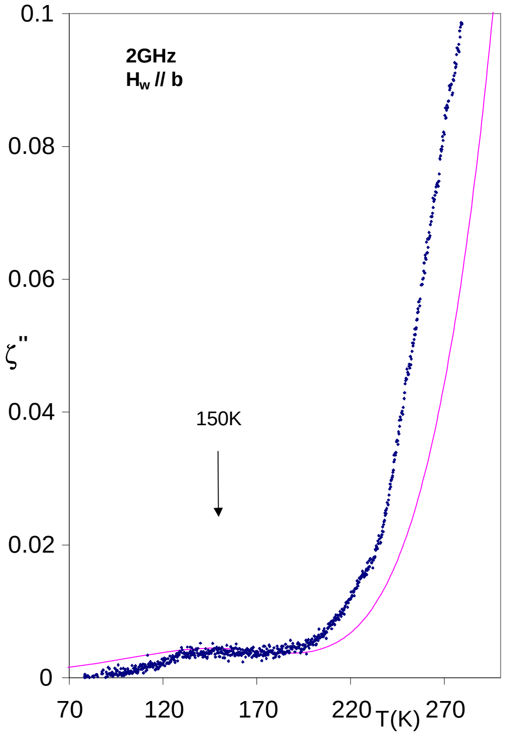

Figure 2: The microwave susceptibility, ζ''_{Hc} at 2 GHz as a function of temperature. Here the dielectric peak loss moved down to 150K at 2 GHz.

chain/ladder compounds of the $Sr-Cu-O$ family, the superconducting cuprates such as $Y:123$ and the $Hg-Ba-Cu-O$ family, and the CMR manganites $La-Sr-Mn-O$. New strong dielectric contributions appear at high frequencies both as dispersion-like changes in ζ'_H accompanied by absorption peaks in ζ''_H below charge ordering transitions.

CONCLUSION

In conclusion we have performed microwave measurements on $La_{5/3}Sr_{1/3}NiO_4$ and observed signatures of charge dynamics which are not observed in other measurements. Our results show that charge ordering is accompanied by the occurrence of dielectric relaxation modes with a large dynamic susceptibility. The present results have important implications for microwave measurements on other oxide materials, such as the spin ladder and superconducting materials, since they demonstrate the phenomenology associated with stripe formation and charge ordering.

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†*email* : *srinivas@neu.edu*.

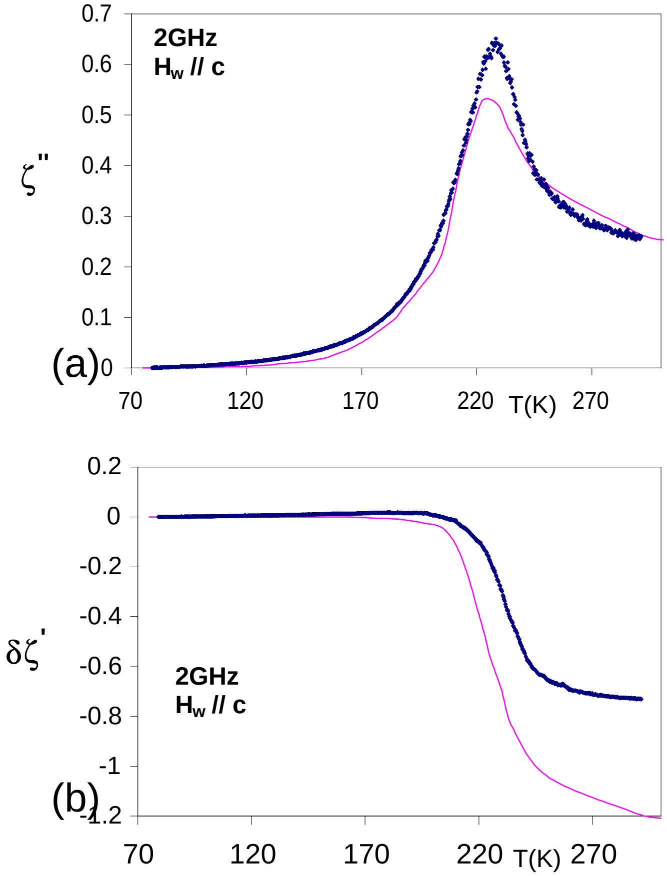

Figure 3: The microwave susceptibility, ζ''_{HC} (a), $\delta\zeta'_{Hc}$ (b) at 2 GHz as a function of tempreture. Here the conductivity term dominates and results in the large peak in ζ''_{HC}. The dielectric loss peak is present but much smaller.

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