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Observation of 300 K high energy magnetodielectric contrast in the bilayer manganite (La$_{0.4}$Pr$_{0.6}$)$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$

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Large high energy magnetodielectric effects are observed in the bilayer manganite (La$_{0.4}$Pr$_{0.6}$)$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$, a direct consequence of exploiting magnetoresistance changes associated with the field driven spin-glass insulator to ferromagnetic metal transition and its high temperature remnant. The low temperature magnetodielectric contrast is as large as ~100% near 0.8 eV at 10 T and over 10 000% in selected phonon regions. The 300 K magnetodielectric contrast is ~20% near 1.1 eV at 30 T. The results are potentially useful for magnetic memory applications away from the dc limit. © 2007 American Institute of Physics. [DOI: 10.1063/1.2757120]

The magnetodielectric (MD) effect, a change in dielectric constant $\varepsilon_1$ with magnetic order or applied magnetic field, is attracting attention due to the fundamental physics that underlies this behavior in complex materials. The static magnetodielectric effect is most commonly investigated, although energy-dependent changes in the dielectric constant are complementary. Static dielectric properties are typically measured in a parallel plate/capacitance geometry at low frequencies (Hertz to Megahertz). Challenges with this technique include contact problems, potential dead layers, edge effects, and distinguishing between leaky dc magnetoresistance and true magnetocapacitance in weakly metallic and/or inhomogeneous systems. A contactless technique, such as optical spectroscopy, eliminates some of these issues. Measurement of the spectroscopic response also provides a way to distinguish between leaky magnetoresistance and true magnetocapacitance because the energies of interest in dielectric spectroscopy are far from the dc limit. At the same time, the electromagnetic spectrum is very broad, and the dielectric constant $\varepsilon_1$ is a strong function of energy. This opens the possibility of exploiting changes in $\varepsilon_1$ over a wide energy range, essentially as a multichannel information storage system. High energy magnetodielectric (HEMD) contrast was recently observed in several materials including inhomogeneously mixed-valent $K_xV_2O_8$ (~5% at 30 T near 1.2 eV), kagome staircase compound $Ni_xV_2O_4$ (~16% at 30 T near 1.3 eV), and hexagonal multiferroic HoMnO$_3$ (~8% at 20 T near 1.8 eV). In these materials, the HEMD effect derives from spin-lattice-charge mixing. Part of our continuing strategy to increase dielectric contrast focuses on exploiting metal-insulator transitions, charge and orbital ordering, and other electronic (beyond ferroelectric) mechanisms that are known to drive significant changes in the optical constants. Here, we report large HEMD contrast in (La$_{0.4}$Pr$_{0.6}$)$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$, behavior that results from a change in magnetoresistance at a magnetic field driven spin-glass insulator to ferromagnetic metal transition. Remnants of the transition also give HEMD contrast at 300 K. (La$_{0.4}$Pr$_{0.6}$)$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$ derives from the La$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$ parent compound, a double-layer perovskite crystallizing in a body-centered tetragonal structure (space group $I4/mmm$). The (La$_{1-x}$Pr$_x$)$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$ series provides an opportunity to investigate the physical properties of bilayer manganites as a function of chemical composition, tuning that gives rise to a rich phase diagram in this and related systems. The parent compound ($z=0$) displays a paramagnetic insulator to ferromagnetic metal transition at $T_c=120$ K. With increasing Pr substitution, $T_c$ decreases, and eventually disappears ($z=0.6$), establishing a spin glasslike insulating ground state. The ferromagnetic metallic state that is suppressed by chemical pressure in (La$_{0.4}$Pr$_{0.6}$)$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$ is recovered under magnetic field. The field-induced transition is between 2 and 5 T, depending on the probe technique, above which the system is in the ferromagnetic metallic state. The local structure around the Mn centers changes dramatically through the critical field.

Single crystals of (La$_{0.4}$Pr$_{0.6}$)$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$ were grown from sintered rods of same nominal composition by the floating-zone technique. Near normal $ab$ plane reflectance of (La$_{0.4}$Pr$_{0.6}$)$_{1.2}$Sr$_{1.8}$Mn$_2$O$_7$ was measured over a wide energy range (3.7 meV–6.5 eV) using a series of different spectrometers and magnets ($H||c$) as described previously. A Kramers-Kronig analysis was employed to obtain the optical constants: $\varepsilon(E)=\varepsilon_1(E)+i\varepsilon_2(E)$. This work focuses on changes in the dispersive response, as measured by $\varepsilon_1(E)$, in a magnetic field. Complementary impedance measurements were carried out at 30 kHz in a two-wire configuration with the ac current parallel to the applied field.

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The high impedance insulating state at 0 T is suppressed with the remnants of the spin-glass insulator to ferromagnetic metal transition in \( \text{La}_{0.4}\text{Pr}_{0.6}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7 \) to achieve a 300 K HEMD effect. Figure 2 displays the room temperature dielectric properties at 0 and 30 T. The dispersive response shifts to lower energy in applied magnetic field, consistent with the previous observation of redshifted oscillator strength in the optical conductivity in the ferromagnetic metallic state.20 Strikingly, \( \Delta \varepsilon_1/\varepsilon_1 \) displays 20% contrast near 1.1 eV (inset of Fig. 2). Although weaker than that at 4.2 K, this finding demonstrates that electronic transitions can be harnessed for 300 K HEMD effects. Reviewing the Tomioka-Tokura global phase diagram in complex perovskites,19 the title compound is sitting in close proximity to the phase boundary and is therefore susceptible to physical tuning—even when the transition itself is no longer well defined. Control of disorder therefore provides an important route for tuning HEMD contrast.19,20

It is useful to compare the HEMD effect in \( \text{La}_{0.4}\text{Pr}_{0.6}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7 \) with static results. Due to the dc conductivity of these single crystal samples (~250 \( \Omega^{-1} \text{ cm}^{-1} \) in the high field phase at 4 K), we investigate the low frequency magnetic response in terms of the impedance rather than \( \varepsilon_1 \).27,28 The inset of Fig. 3 displays the low frequency (30 kHz) impedance as a function of magnetic field at 4.2 K. The high impedance insulating state at 0 T is suppressed

FIG. 1. (Color online) (a) Dielectric response of \( \text{La}_{0.4}\text{Pr}_{0.6}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7 \) for \( H=0 \) (solid line) and 10 T (dashed line) \( (\varepsilon'(\omega)) \) at 4.2 K, as determined by Kramers-Kronig analysis of the measured reflectance. (b) The high energy dielectric contrast \( \Delta \varepsilon_1/\varepsilon_1 \) between \( H=10 \) and 0 T at 4.2 K \( (H||c) \).

FIG. 2. (Color online) Dielectric response of \( \text{La}_{0.4}\text{Pr}_{0.6}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7 \) for \( H=0 \) (solid line) and 30 T (dashed line) \( (\varepsilon'(\omega)) \) at 300 K, as determined by Kramers-Kronig analysis of the measured reflectance. The inset displays the high energy dielectric contrast \( \Delta \varepsilon_1/\varepsilon_1 \) between \( H=30 \) and 0 T at 300 K \( (H||c) \).

FIG. 3. (Color online) Magnetoimpedance contrast of \( \text{La}_{0.4}\text{Pr}_{0.6}\text{Sr}_{1.8}\text{Mn}_2\text{O}_7 \) as a function of magnetic field \( (H||c) \) at 300 K. The inset displays the magnitude of the impedance as a function of magnetic field \( (H||c) \) at 4.2 K. The sample was zero-field cooled.
with increasing magnetic field, leading to a large change in impedance between 0 and 5 T. Due to the first order nature of the transition, the sample remains trapped in the metastable low impedance state as $H$ is decreased toward 0 T. Field-induced transitions and trapping in a metastable state have been observed in resistivity measurements of (La$_{0.4}$Pr$_{0.6}$)$_2$Sr$_{1.8}$Mn$_2$O$_7$ and were attributed to the strong magnetoresistive coupling in this system. This static spin-charge coupling persists to higher temperatures. The main panel of Fig. 3 displays the 300 K magnetoimpedance contrast as a function of magnetic field. The impedance of the sample changes up to 2.5% in a 5 T field. Similar magnetoimpedance has been observed close to the ferromagnetic ordering temperature in other manganites and was attributed to the dependence of the skin depth on both magnetic field and frequency. Clearly, these room temperature effects may also support high energy magnetodielectric contrast, a direct consequence of magnetic field. Other electronic mechanisms in complex oxides may also contribute to the skin depth on both magnetic field and frequency. Clearly, these room temperature effects represent significant spin-charge coupling persisting to temperatures well above the magnetic transition temperature, in line with the 300 K HEMD effect.

To summarize, we observed high energy magnetodielectric contrast in the bilayer manganite (La$_{0.4}$Pr$_{0.6}$)$_2$Sr$_{1.8}$Mn$_2$O$_7$, a direct consequence of magnetoresistance changes at the field driven spin-glass insulator to ferromagnetic metal transition. The remnants of this transition can be used to achieve dielectric contrast at room temperature. Other electronic mechanisms in complex oxides may also support high energy magnetodielectric contrast, eliminating the practical need to identify high temperature multiferroics.

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