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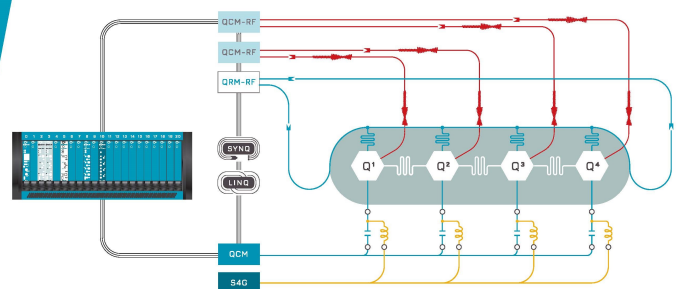Applied Physics Letters **93**, 212105 (2008); https://doi.org/10.1063/1.3036532

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Strong resistance nonlinearity and third harmonic generation in the unipolar resistance switching of NiO thin films

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We investigated third harmonic generation in NiO thin films that exhibit unipolar resistance switching. We found that low resistance states (LRSs) were strongly nonlinear with variations in the resistance R as large as 60%, which was most likely caused by the Joule heating of conducting filaments inside the films. By carefully controlling the applied dc bias, we obtained several LRSs, whose values of the third harmonic coefficient B_{3f} were proportional to R^{2+w} ($w = 2.07 \pm 0.26$). This suggested that the resistance changes in the NiO films were accompanied by connectivity changes of the conducting filaments, as observed in classical percolating systems. © 2008 American Institute of Physics. [DOI: 10.1063/1.3050519]

Unipolar resistance switching (RS) occurs when two reversible resistance states exist under one bias voltage polarity.[1] This intriguing phenomenon has been observed in numerous materials including many binary oxides.[2] Recently, unipolar RS has attracted a great deal of interest partly because of its technological potential for nonvolatile memory.[2,3] Although its physical mechanism is still a matter of debate, it is widely accepted that it occurs due to the formation and rupture of conducting filaments under an external bias voltage.[2,4–10]

The cluster structure properties of the conducting filaments, such as connectivity and topology, are likely to play a very important role in unipolar RS.[8] However, to date there have been no serious attempts to investigate these properties. By examining the conducting spots on a sample surface, conductive atomic force microscopy can probe the appearance and disappearance of percolating conduction filaments but not how they are connected inside the bulk sample.[8] It is very difficult to investigate the cluster structure of the filaments using other microscopic tools such as transmission electron microscopy[7] because the filaments form very complicated and winding channels inside a three-dimensional sample. Therefore, our understanding of the cluster structure of conducting filaments in real RS materials remains relatively poor.

Here, we present an investigation of the nonlinear electrical responses of NiO thin films using third harmonic generation (THG) techniques. Our NiO films showed unipolar RS and very strong nonlinear electrical responses due to the Joule heating. We compared THG signals during multilevel switching processes and found that they scaled with resistance. These observations suggested that the RS process was accompanied by changes in connectivity among the conducting filaments. We also compared the critical exponents of the THG signals with those of classical percolating systems.

We grew polycrystalline NiO thin films on $Pt/Ti/SiO_2/Si$ substrates using dc magnetron reactive sputtering. The detailed fabrication methods for NiO thin films and Pt top electrodes were described elsewhere.[1] We measured the dc current-voltage (I_{dc}-V_{dc}) curves using a semiconductor parameter analyzer (Agilent 4155C, Agilent Technologies, Santa Clara, CA).

Figure 1(a) shows the I_{dc}-V_{dc} curve of a Pt/NiO/Pt capacitor that exhibited characteristic unipolar RS. The NiO thin film was highly insulating in the pristine state. When we applied a V_{dc} of 3–5 V (not shown here), I_{dc} increased suddenly, which could have caused a dielectric breakdown. To prevent damage, we kept the current in the NiO film below the compliance level I_{comp}. Immediately after this forming process, the film entered a low resistance state (LRS). When V_{dc} was increased above 0.5 V, the film changed from the LRS to a high resistance state (HRS); we called this the reset process. As we increased V_{dc} again in the HRS, the film changed back into the LRS for V_{dc} between 1.5 and 3.0 V; we called this the set process. For both forming and set processes, we used an I_{comp} of 1 mA.

By carefully controlling the reset voltage, we could observe multilevel reset processes [Fig. 1(b)]. Just after forming, our NiO film was in the LRS, denoted as "1" in Fig. 1(b). An abrupt resistance change was observed as we increased V_{dc} slowly to 0.49 V. Note that the film was not changed into the HRS but into another level of LRS, indicated by "2." By carefully controlling V_{dc}, we obtained four intermediate LRS levels. When V_{dc} was increased above 0.56 V, the sample finally changed into the HRS.

The I_{dc}-V_{dc} curves in the LRS showed strongly nonlinear variation with resistance. As shown in Fig. 1(a), the film deviated strongly from the Ohmic behavior (dashed line), particularly near the reset voltage. As shown in the inset of Fig. 1(a), the nonlinearity of the film resistance R_{dc} was 60%. As shown in the inset of Fig. 1(b), such nonlinearity persisted at all levels of the LRS. It is likely that the increase in
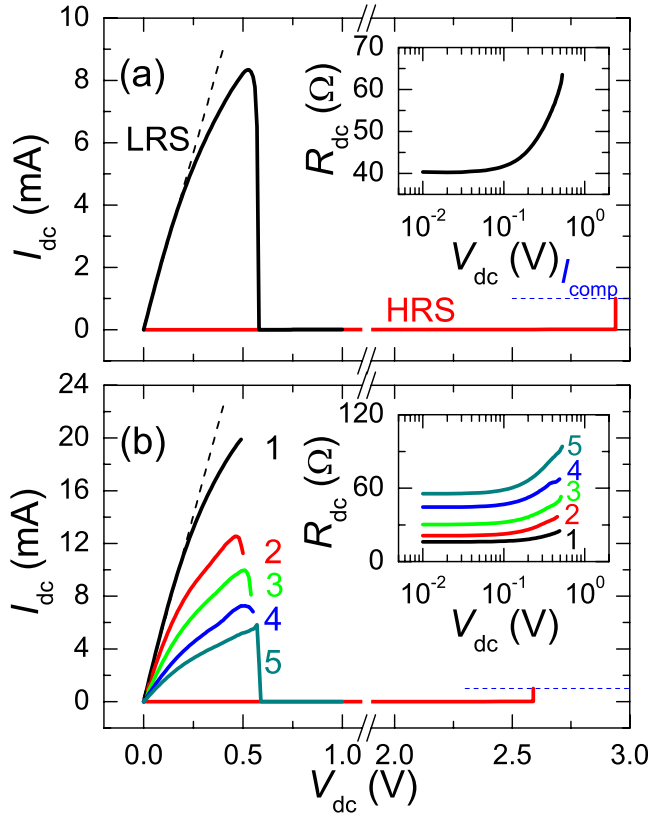
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FIG. 1. (Color online) (a) Characteristic dc current-voltage (I_{dc}-V_{dc}) curve of NiO films showing unipolar RS. The LRS had a nonlinear I_{dc}-V_{dc} curve that deviated from the Ohmic behavior (dashed line). (b) Changes in the I_{dc}-V_{dc} curves of the NiO film during multilevel switching. For (a) and (b), we set the compliance current I_{comp} to 1 mA to avoid permanent dielectric breakdown. The insets show the strong nonlinear changes in dc resistance $R_{dc}(\equiv V_{dc}/I_{dc})$ for the NiO film.

R_{dc} was derived from the temperature increase of conducting filaments due to the Joule heating.

To obtain further insight, we investigated the THG responses due to the Joule heating.[11–14] Suppose that a current i flows through a conducting filament with resistance r. Due to the Joule heating effects, r increases by $\Delta r = r_o\beta\Delta T$, where r_o and β are the zero-biased value and temperature coefficient of resistance for the conducting filament, respectively. The filament's temperature T rises due to the Joule heating, so ΔT is proportional to ri^2. With $\Delta r \propto r_o ri^2$, the voltage drop v across the filament is $v \approx r_o i + K r_o r i^3$, where K is a proportionality coefficient. When an ac current with a frequency f $i = i_{ac}\cos(2\pi ft)$ flows through the resistor, $v(t) = r_o i_{ac}\cos(2\pi ft) + v_{3f}\cos(6\pi ft)$, where $v_{3f} = (i_{ac}\Delta r)/2$. Hence, by measuring the $3f$ component, we could investigate the resistance nonlinear response due to the Joule heating.

We performed THG experiments by placing a NiO film in one arm of a Wheatstone bridge circuit. We supplied an ac voltage V_{ac} generated by a lock-in amplifier (SR830, Stanford Research Systems, Sunnyvale, CA). We measured the $3f$ voltage component V_{3f}. We could also obtain R for the NiO film independently by balancing the Wheatstone bridge circuit. Using the measured R values, we determined $I_{ac} = V_{ac}/R$. We obtained most V_{3f} data at $f = 10$ Hz and room temperature. We also performed THG measurements at other f values (i.e., 1 Hz–2 kHz). For a given value of V_{ac}, we found that V_{3f} was almost independent of f.[11,12]
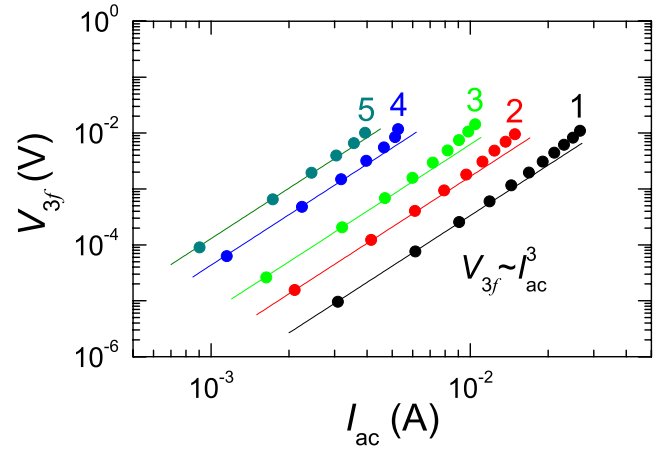

FIG. 2. (Color online) Third harmonic voltages V_{3f} for the five levels of LRS shown in Fig. 1(b). At all levels, V_{3f} was proportional to I_{ac}^3 at low I_{ac} (solid lines). The relation $V_{3f} \propto I_{ac}^3$ suggested that the third harmonic signals could have been caused by the Joule heating.

Figure 2 shows the behavior of V_{3f} for different levels of the LRS. As the level number of the LRS increased, V_{3f} also increased significantly. For a given value of I_{ac}, e.g., 3 mA, the V_{3f} value could vary by a factor of around 300. This variation was much larger than the R variation, which was around 3.5, indicating that V_{3f} was very sensitive to the nonlinear response of the NiO film. As shown by the solid lines in Fig. 2, V_{3f} was proportional to I_{ac}^3 at low I_{ac} of all LRS levels. This was consistent with our earlier arguments based on THG due to the Joule heating. For later use, we defined the third harmonic coefficient B_{3f} as V_{3f}/I_{ac}^3.

Careful analyses of B_{3f} can provide new insights into changes in the cluster structure parameters, such as connectivity, of the conducting filaments during the reset process.[11–14] For an inhomogeneous medium, the current distribution should not be uniform. We assumed that the response of the NiO films could be represented by a network with randomly distributed conducting filaments. While R measures the second moments of the current distribution, B_{3f} probes the fourth moments[11–14]

$$\frac{B_{3f}}{R^2} \propto \frac{\sum i_b^4}{\left(\sum i_b^2\right)^2}, \tag{1}$$

where i_b is the current flowing through each conducting filament b. If there is no change in the connectivity of filaments, the right term in Eq. (1) remains constant and B_{3f} is proportional to R^2. As shown in Fig. 3(a), for each LRS, $B_{3f} \propto R^2$. This experimental result indicated that there was no change in the connectivity of conducting filaments while the LRS level was maintained.

When the connectivity of the filaments changes, the R dependence of B_{3f} varies. Traditionally, electrical breakdown in semicontinuous metal films has been treated using percolation theory.[11–15] The configuration change in the connectivity of conducting components changes the current distributions [i.e., in Eq. (1)], leading to $B_{3f}/R^2 \propto R^w$.[11–14] According to classical lattice percolation theory,[16] $0.82 < w < 1.05$. For two-dimensional semicontinuous metal films,[12,13] it has been reported that $1.2 < w < 2.0$.

Figure 3(b) shows the dependence of B_{3f} on R for a given V_{ac} value. The open triangles and squares indicate the
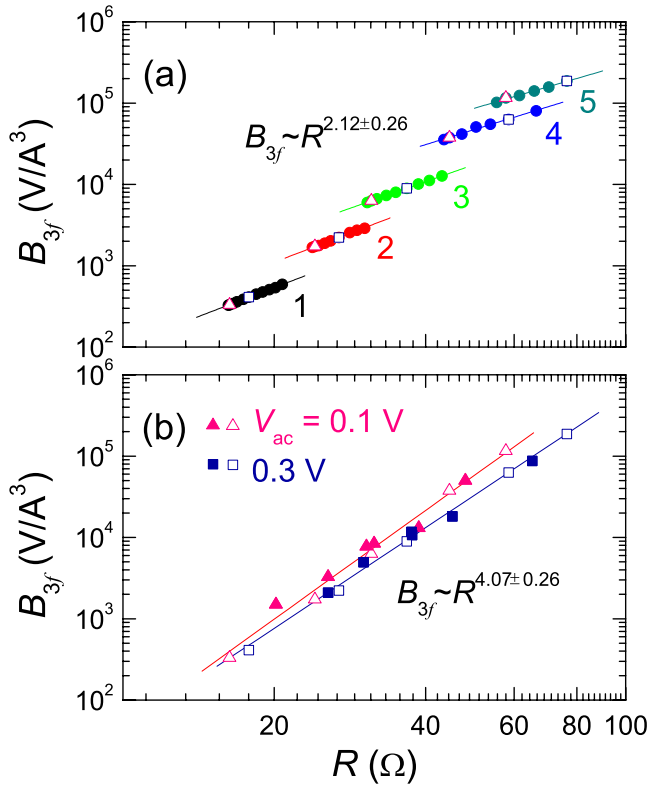
FIG. 3. (Color online) Dependence of the third harmonic coefficient B_{3f} on R for (a) each level of LRS and (b) a given value of V_{ac}. The open triangles and squares in (b) correspond to the data shown by the open symbols in (a). We repeated the same experiment with other NiO films and plotted the results using solid symbols. Note that $B_{3f} \propto R^{w+2}$ with (a) $w \approx 0$ and (b) $w = 2.07 \pm 0.26$.

B_{3f} values at $V_{ac} = 0.1$ and 0.3 V, respectively, for the five levels of LRS in Fig. 1(b). To test the reliability of our data, we repeated the THG experiments with other NiO capacitors. These results are indicated with the solid symbols. We found that during the multilevel reset processes, $B_{3f}/R^2 \propto R^w$ with $w = 2.07 \pm 0.26$. The nonzero value of w indicates that the connectivity of the conducting filaments changed for every intermediate LRS level during the multilevel reset processes. The w-value was similar to that of ion milled Au films ($w = 2.0 \pm 0.1$).[17]

We analyzed the data at different V_{ac} and f and found that the w-value remained the same. We also performed THG measurements at different temperatures. Although the values of B_{3f} at 10 K became larger by a factor of 10 than those at room temperature, the w-value was almost the same. These observations indicated that the w-value did not depend on experimental variables such as V_{ac}, f, or measurement temperature. As in classical percolation problems, this critical exponent may provide a new way to study the cluster structure of the conducting filaments inside unipolar RS material

systems. There have been no reports of theoretical studies on the w-value of unipolar RS materials. Further studies on this critical exponent are therefore necessary.

In summary, we found that the NiO thin films exhibited RS behavior with strong nonlinear electric responses. Using THG, we demonstrated that the nonlinear resistance responses were most likely caused by the Joule heating effects. We found that the third harmonic signals of multilevel LRSs scaled with resistance with a critical exponent value, as in some percolating systems. The results presented here suggest that THG could be an effective indicator of how conducting filaments are connected and disconnected in unipolar RS materials.

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