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A Kondo insulating memristor

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We report memristor behavior in a macroscopic bulk Kondo insulator $Ce_3Bi_4Pt_3$ below 10 K. The ac current and voltage relation characteristic of memristors, pinched hysteresis loops which merge into a single line at high frequency, are systematically observed. A standard lock-in technique is used to probe the origin of this exotic behavior which cannot be explained solely by a static resistance drop arising from joule heating. Rather the response appears to be associated with the formation of dynamic thermal impedance in the dissipative regime. © *2012 American Institute of Physics*. [http://dx.doi.org/10.1063/1.4733328]

In circuit theory, the four fundamental variables of voltage, current, magnetic flux, and charge have physical relationships linking pairs of the four except in the case of magnetic flux and charge. Defining magnetic flux and charge mathematically via time integrals of voltage and current, respectively, the missing hypothetical circuit element is known as a memristor[1] with the variables flux and charge linked by a constitutive relation. In 2-terminal linear circuit elements, memristance is not state dependent but a constant converging into pure resistance, but 2-terminal nonlinear circuit elements can have memory resistance controlled by one of the two mathematical state variables. The finger print of a memristor is a pinched hysteresis loop in the ac current-voltage (IV) response which merges into a single line when the ac driving frequency goes to infinity.[2] Titanium dioxide has been a focus of memristor research in a nano-crossing bar structure,[3] and similar nano-junction devices have also been demonstrated.[4–6] Nano-structure is not a necessary condition for the realization of memristors: any nonlinear system satisfying an autocatalytic relation[2,7] is a possible candidate for a memristor even in macroscopic bulk materials. A central issue for overcoming the problem of device aging is to find a mechanism other than ionic migration. The heavy fermion compound $Ce_3Bi_4Pt_3$ is a so-called Kondo insulator[8] with a hybridization gap[9] which opens below approximately 100 K. This material has been studied via various experimental methods.[10–14] The electrical resistivity measured at low current shows more than 2 orders of magnitude increase from 300 K to 2 K characterizing a continuous metal insulator transition, as shown in Fig. 1. The resistance value increases from 2.4 mΩ to 3.3 Ω as the temperature decreases from 300 K to 2 K in a $1 \times 2 \times 0.5\ \mathrm{mm}^3$ size sample, and it is still a relatively good conductor at 2 K. The single crystals of $Ce_3Bi_4Pt_3$ used in the measurements reported here were grown from $CePtBi_5$ in quartz encapsulated alumina crucibles cooled at 12 °C/h from 1050 °C to 400 °C, at which temperature, the crystals were separated from the remaining molten flux by centrifuging through a quartz wool plug.

A series of pinched hysteresis loops are shown in Fig. 2 measured with a 4-probe synchronous sinusoidal ac current source and ac voltage acquisition on a $Ce_3Bi_4Pt_3$ single crystal at 2 K. Unlike typical Lissajous curve acquisition with an oscilloscope, the synchronous data generation and acquisition are critical for revealing memristive behavior in dissipative systems. The synchronous method used records the ac IV relation as it evolves with cycling. Silver epoxy and annealed Pt wires are used to make ohmic contacts. The sample was mounted on the sample puck in a Quantum Design PPMS (Physical Property Measurement System) using cryogenic thermal grease with 10^{-6} Torr vacuum in the surrounding chamber. The dashed lines in Fig. 2 show the first Lissajous cycle curve for each frequency. The main characteristic of memristors is seen in the Lissajous curves which show a clear evolution of pinched hysteresis with frequency merging into a single line at 10 kHz. Unlike the binary oxide systems, the pinched loops of $Ce_3Bi_4Pt_3$ exhibit no self-crossing[15] or non-symmetric geometry due to self heating.

A similar time evolution of the Lissajous curves at a fixed high frequency has been observed in an ionic migration system and demonstrated as well in the spike timing dependent plasticity of biological synapses.[5] A memristor is an ac driven device, and our slow dc IV measurements with 5 s delay before each data point acquisition do not display the hysteresis, as is clear from the inset of Fig. 3.

In view of the low resistance value of the sample even at 2 K, the shape of the hysteresis curve and its time evolution are unexpected within the framework of a static resistive model or the static resistance drop caused by joule heating. For instance, current ramping from 20 to 0 mA does not follow the same trajectory as ramping from 0 to 20 mA, but instead behaves as if there is considerable time lag between the source current and the measured voltage. At the low frequencies of 1 and 5 Hz, the first and the final states are the same and exactly overlap. However, with increasing frequency, the ac electrical responses do not overlap but exhibit a time evolution of the Lissajous curves with conspicuous hysteresis. The derivative of the initial lines, which correspond to the resistance of the sample, has the same value: 3.3 Ω. The frequency dependence can be explained by joule self heating. At 1 Hz, the sample starts at zero current at the temperature of the base plate, 2 K. As the current through the sample increases from 0 to 20 mA, the sample temperature increases as the current increases due to joule heating. When the current is cycled back to zero the sample temperature recovers the initial value of 2 K. The unusual impedance making the hysteresis occurs during the heating and cooling

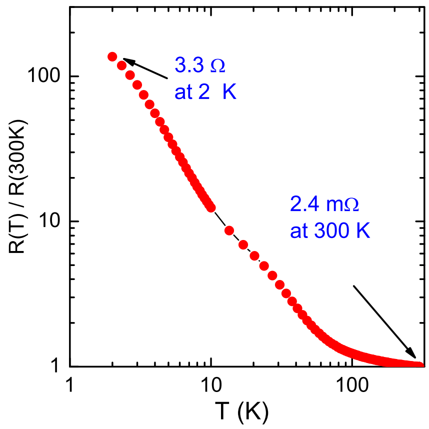

FIG. 1. Temperature dependence of normalized resistance of single crystal $Ce_3Bi_4Pt_3$ from 300 K to 2 K

cycle. The symmetry in the first and third quadrants seen in Fig. 2 reflects this. This tendency persists up to several Hz. With increasing driving frequency, the symmetry of heating and cooling is lost. At 1 kHz, the initial resistance when the current starts flowing is 3.3 Ω, indicating that the sample is at 2 K. Then higher overall resistance (even though it decreases as the current increases) persists to 20 mA for the first cycle, as shown in the 1 kHz plot with the unusual impedance. When the current returns to zero, the sample is not at the initial temperature but at a higher temperature than 2 K. This thermal cycling leads to a non-symmetric evolution of the Lissajous plots at high frequencies. The heating process continues through the following intermediate cycles until reaching a final state. The Lissajous plots for the intermittent states are not symmetric but the final state is symmetric. To clarify the onset of the unusual impedance, we made a standard 4 probe lock-in measurement using the same sample configuration. A small ac current (10 μA and 17 Hz) is superimposed on the much larger dc current and the dc current is ramped to ±20 mA to measure dV/dI and the phase shift of the ac signal. The upper panel of Fig. 3 shows the dV/dI curve obtained simultaneously with the dc IV curve. As the IV slope saturates, its derivative approaches zero. The essential point is that the drop of the measured dV/dI is not simply due to a static resistance change. The lower panel of Fig. 3 shows the phase shift of the small ac signal during the dc current ramping. The lock-in phase tuning is done initially at zero dc current. As the dc current increases, the relative phase of the sample signal voltage does not hold its original value, showing instead a monotonically increasing phase shift. This means that the dynamics of the system cannot be explained within the framework of a static resistive model, but rather a virtual capacitance arises leading to a phase shift between the ac current and voltage. This capacitance produces the memristive pinched hysteresis loop. With increasing frequency, as seen in Fig. 2, the final state resistance becomes smaller and nearly saturates between 1 kHz and 10 kHz into a merged single line. At 10 kHz, where an almost ohmic IV relation is seen, the first cycle takes only 100 μs. These two facts suggest that the memristor behavior is correlated with thermal relaxation. Thus, the pinched hysteresis loops in Fig. 2 reflect development of electrical thermal impedance. The energy balance of the system can be described by generated energy (ΔP) = stored energy (C d(ΔT)/dt) + dissipative energy (D ΔT), where ΔT is the temperature variation corresponding to power variation, ΔP, C, the specific heat and D, the dissipation constant. With the temperature coefficient given by $\alpha = \Delta R/R\Delta T$, the above energy relation leads to the general form of frequency dependent impedance for a sinusoidal perturbation as

$$R = 1 \Bigg/ \left(\frac{D - \alpha P}{2\alpha PR}\right) + i\omega\left(\frac{C}{2\alpha PR}\right). \quad (1)$$

The above impedance converges to resistance R when there is negligible power generation from self heating and has meaning only when it goes beyond the ohmic regime. At zero frequency, the virtual capacitance term has no meaning and the hysteresis in the first and third quadrant disappears. In contrast, increasing frequency leads to the hysteresis phase shift seen in the small signal dV/dI relation. The merging to a single line in the first cycle means that the first cycle occurs without significant heating or thermal impedance, but

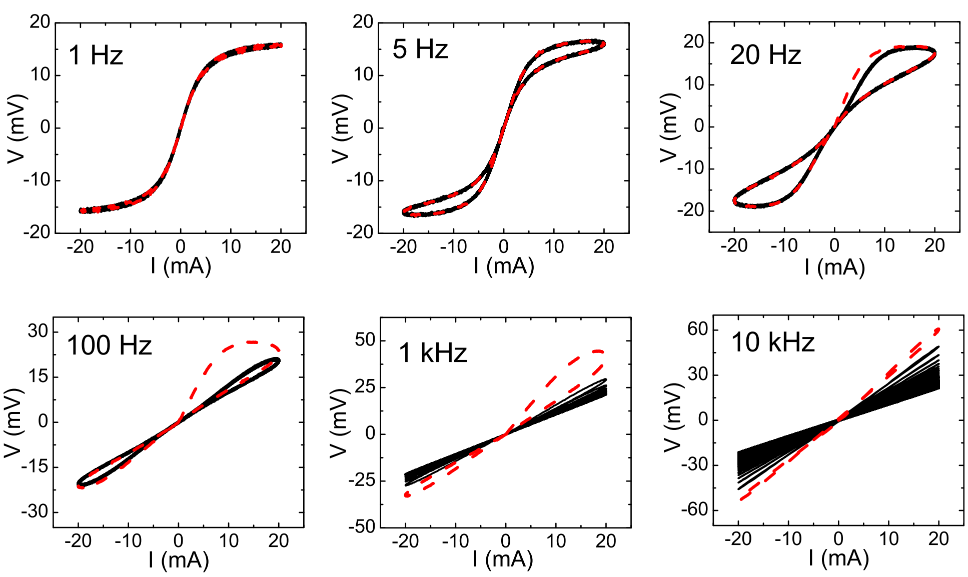

FIG. 2. Frequency dependence of Lissajous curves. As a finger print of memristor, pinched hysteresis and evolution to a merged single line at high frequency are clearly seen.

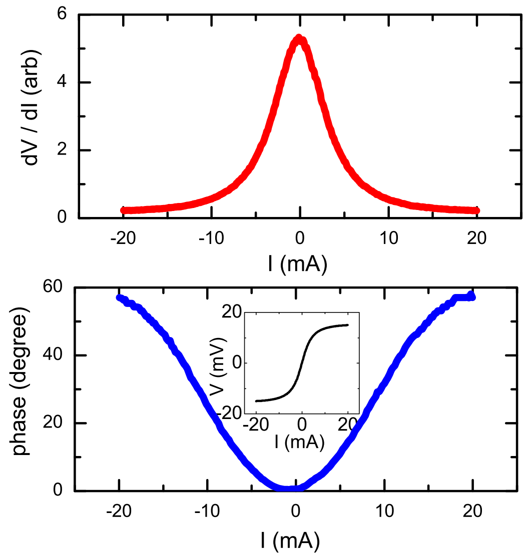

FIG. 3. The upper panel is dV/dI data simultaneously acquired with IV curve shown in the inset from a standard lockin technique. The lower panel is the phase shift of small ac modulation voltage over large dc current ramping.

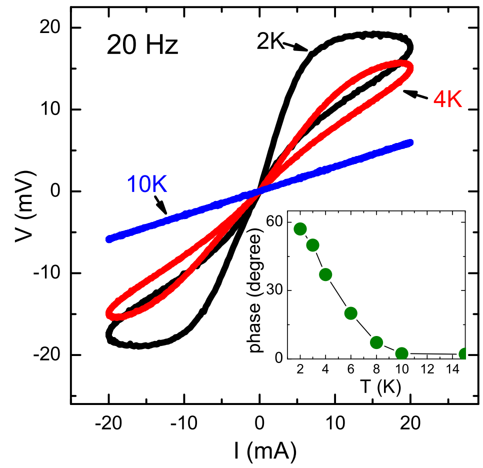

FIG. 4. Temperature dependence of Lissajous curve at 20 Hz. The inset shows the temperature dependence of phase shift of acquired ac voltage against the ac modulation current.

the resistance drops with each successive cycle and after many cycles produces the linearity in the IV relation. We can confirm the presence of the thermal impedance by changing the sample base temperature, as shown in Fig. 4.

The inset shows the phase shift dependence with temperature at 20 mA. Starting at 2 K, the sample self heats and generates a 57° phase shift at 20 mA, indicating a current dependent thermal capacitance. This capacitance leads to the pinched hysteresis curve in Fig. 4 for 2 K and 20 Hz. As the base temperature increases, the sample resistance drops rapidly and the heat generation becomes smaller. The phase shift variation with base temperature range change 2 K to 15 K shows a clear evolution. The sample resistance becomes smaller than 300 mΩ above 10 K, and the self heating effect becomes negligible so that almost zero phase shift is observed and no hysteresis is observed in the Lissajous curve.

In summary, we have shown that a Kondo insulator $Ce_3Bi_4Pt_3$ satisfies the necessary condition for a memristor, exhibiting pinched hysteresis in its Lissajous IV curves, which merge at high frequency into a single line. We interpret the origin of this memristive behavior as a virtual thermal impedance arising from self heating. Its origin is quite different from other binary oxide systems where ionic migration, which eventually degrades the performance, dominates. Typically, Kondo insulators show resistance increasing below room temperature, each with its own individual temperature characteristic. Our thermal model may suggest new memristors in contemporary silicon on insulator (SOI) technology where such self heating leads to nonlinear behavior.[16–18]

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