Electronic transport properties of $Fe(qsal)_2(TCNQ)_2$ molecular films

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Motivation

Microelectronics applications in the GHz range

- Conventional memory devices are energy inefficient and rapidly increasing in demand
- Energy use of devices must be greatly decreased to avoid crisis
- Size and efficiency improvements to conventional transistor mechanisms encounter physical, technological and financial limits

Transistors utilizing molecular magnets could dramatically decrease energy costs

- Decreases transistors per memory device from 6 to 1
- Allows for nonvolatile devices (only energy cost for read/write operations)
- Energy cost < 5 aJ
- Need to determine estimate read/write speed - lacks application if not fast enough

Switching behavior derived from high-frequency impedance

- Cannot directly measure at frequencies in theoretical operating range

[1] T.K. Ekanayaka, R. Streubel, P.A. Dowben et al., Mater. Chem. Phys. 296, 127276 (2023).

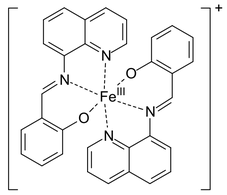
[2] P. A. Dowben, D. E. Nikonov, A. Marshall, and C. Binek, Appl. Phys. Lett. 116, 080502 (2020).

[3] K. Ekanayaka, G. Hao, A. Mosey, A. S. Dale, X. Jiang, A. J. Yost, K. R. Sapkota, G. T. Wang, J. Zhang, A. T. N'Diaye, A. Marshall, R. Cheng, A. Naeemi, X. Xu, and P. A. Dowben, Magnetochemistry 7, 37 (2021).

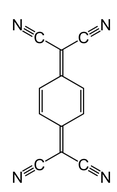
Thin Film Preparation

Molecular magnet solution deposited on gold electrode substrate

- Sample deposited from solution via drop casting
- Current splits into many paths, flows through 10 μm of sample
- Current-voltage (I-V) curves dramatically change at unexpectedly low currents, with repeated/sustained measurements causing a transition from non-ohmic to ohmic behavior
- Conductance greatly increases after transition
- Transition could be from burning or crystalline change and likely alters molecular magnet behavior
- System modelled as a resistor and capacitor in parallel, in series with contact resistance

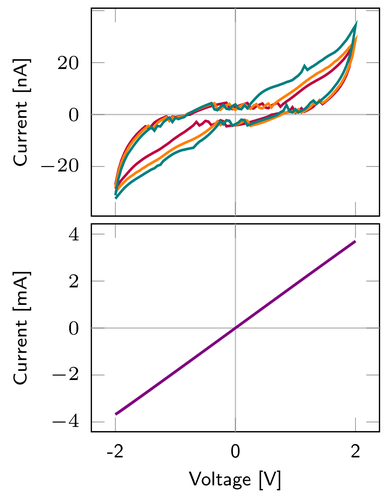


$[Fe(qsal)_2]^+$



TCNQ

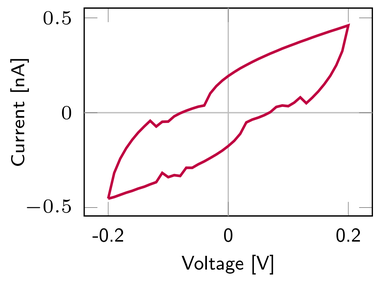
Optical image



DC Current-Voltage Behavior

Consistent non-ohmic behavior for decreased voltage ranges

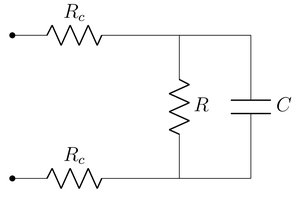
- Small in-plane voltages (< 0.5 V) required to avoid sample degradation
- Lower conductance than expected
- Significant remanent current at 0 V which depends greatly on voltage range
- Remanent current could result from both capacitive behavior (discharging) or leakage current, which motivates characterization of the time evolution at different voltages

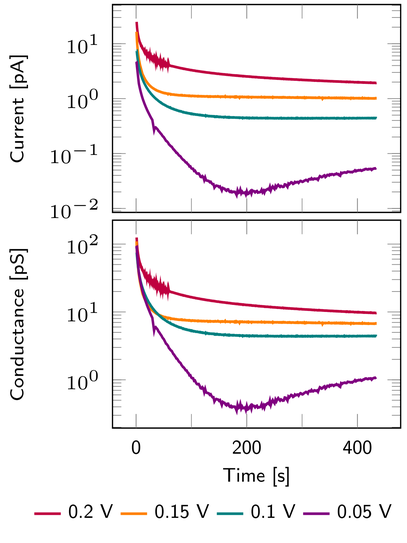


Time-dependence of Current

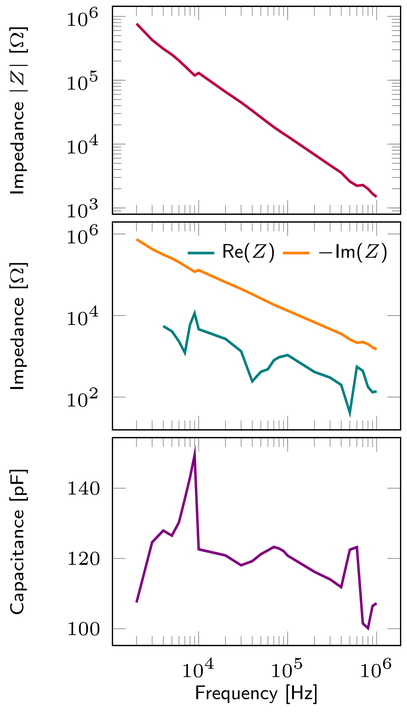
Atypical RC charging behavior

- Held at a constant voltage and measured time evolution of current
- Non-exponential decay, with varying decay shapes at different voltages, indicating behavior deviating from in-series RC circuit model
- Conductance curves and asymptotic values do not align, suggesting degradation or non-linear leakage
- The presence of significant non-capacitive behavior/leakage justifies circuit model
- Sample transitioned to linear I-V behavior after final measurement (0.05 V), indicating degradation from sustained current





AC Behavior



Well-behaving with low impedance at high frequencies

- Frequency-dependence measurements taken from low to high frequency
- Extrapolated trend from low impedance in MHz range predicts favorable results in GHz range
- No noticeable inductive behavior for high frequencies
- Leakage current should only be significant for smaller frequencies
- Circuit model decently explains decays of effective resistance and capacitive reactance with increasing frequency, though effective resistance decays more slowly than expected

$R \approx 40\ \mathrm{M\Omega} \quad C \approx 120\ \mathrm{pF}$

Characteristic frequency

$\omega_0 = \frac{1}{RC} \approx 200\ \mathrm{Hz}$

Effective resistance

$\mathrm{Re}(Z) = \frac{R}{1+\frac{\omega^2}{\omega_0^2}} + 2R_c$

Capacitive reactance

$\mathrm{Im}(Z) = \frac{-R}{\frac{\omega_0}{\omega}+\frac{\omega}{\omega_0}}$

Technological application possible if degradation is prevented

- Issues seem largely due to the films rather than the specific molecules used, increasing film quality could prevent degradation
- Scanning electron microscope images of samples are needed, both before and after the transition in behavior
- Circuit model could be confirmed with low frequency measurements

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