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Interface superconductivity in graphite- and CuCl-based heterostructures

**Yakov Kopelevich
UNIVERSIDADE EADUAL DE CAMPINAS**

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Final Report**

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The results obtained within the framework of this project shed a new light on the long-standing problem of possible high- temperature superconductivity in CuCl. The obtained experimental evidence indicates that the low resistance state in CuCl is governed by motion of charge density wave(s) along quasi-one-dimensional filaments. The electric-field-induced metallic state, non-linear current-voltage characteristics, intermittent switching between low- and high-resistance states, aging effect, and negative parabolic magneto-resistance, all these can be understood assuming the Fröhlich-type (super)conductivity in the studied material.

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Final Performance Report

Principal Investigator: Dr. Iakov Kopelevitch

Instituto de Física “Gleb Wataghin”, Universidade Estadual de Campinas-UNICAMP.
Adress: Rua Sergio Buarque de Holanda, 777, cidade Universitária Zeferino Vaz,
Campinas-São Paulo-Brazil (zip code: 13083-859)

Grant/Contract Number: FA9550-13-1-0056

Program Manager: Dr. Brett Pokines, AFOSR/IO

Grant/Contract Title: *“Interface Superconductivity in Graphite-and CuCl-based Heterostructures”*.

Within the framework of this project, we concentrated on a possible occurrence of high-temperature superconductivity in CuCl.

In what follows, we describe our main achievements.

Copper chloride (CuCl) possesses the zinc blende crystalline structure and has a white color. At normal pressure, CuCl behaves as an insulator with the energy gap of ~ 4 eV.

While CuCl was studied for decades [1-3], its physical properties are still not fully understood. Thus, CuCl demonstrates superdiamagnetism with the susceptibility $\chi \sim -0.8$, accompanied by a resistance drop of about six orders of magnitude, resembling the behavior of superconductors.

This high-to-low resistance state transition can be induced applying either pressure [1-3] or sufficiently high electric field [4, 5]. This insulator-metal transition (IMT) was attributed to the formation of metallic copper filaments within the bulk material. Despite of the observation of IMT in CuCl, the origin of the low-resistance state (LRS) is largely unknown. This motivate us to study LRS in more details.

The samples were prepared pressing the commercially available (Sigma Aldrich and Alfa Aesar) CuCl powder into cylindrical pellets of 5 mm diameter (D) with the thickness of 1 mm. The pressure of 1 GPa was applied using a hydraulic press and a steel pastillator. During the measurements, the samples were kept in an inert atmosphere of He gas inside of commercial cryostats from Janis and Quantum Design (PPMS) Co. The equipment allows the variation of the temperature (T) and magnetic field (B) in the intervals: $2\text{ K} < T < 300\text{ K}$ and $-9\text{ T} < B < 9\text{ T}$.

Two-probe electrical transport measurements were performed with the silver paste (Ag) electrodes placed on upper and lower faces of the sample, as shown in Fig. 1(b). Current-voltage (I - V) characteristics and the resistance vs. time $R(t)$ measurements were performed in the voltage-generator mode. The resistance temperature and magnetic field dependencies $R(T, B)$ were recorded at constant applied current. i. e. in the current-generator mode, Fig. 1(a).

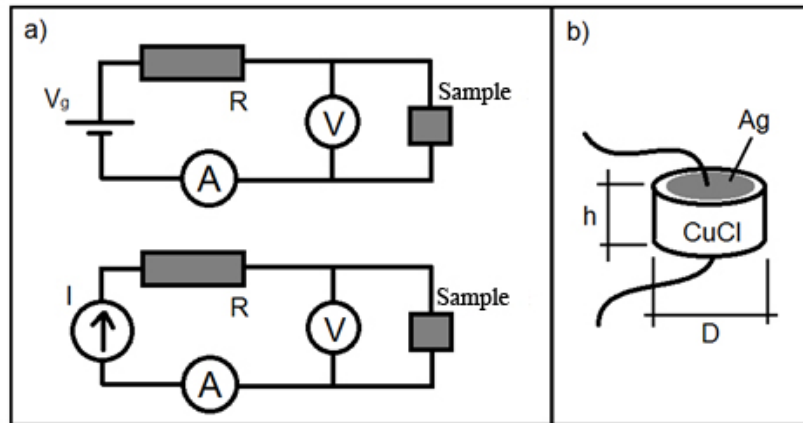


Fig. 1. (a) Voltage- (top) and current-generator (bottom) circuits used in the measurements; (b) Cartoon of measured CuCl samples with diameter (D) and thickness (h).

All our samples were characterized by means of monitoring the resistance as a function of time at a fixed applied voltage. After a certain time delay, typically ranging from 2 to 30 minutes, the samples revealed a sharp reduction of their resistance by 2 - 4 orders of magnitude, depending on the sample details (Fig.2, left panel).

No transition to the low-resistance-state was observed for oxidized (greenish) phase (Fig. 2, right panel).

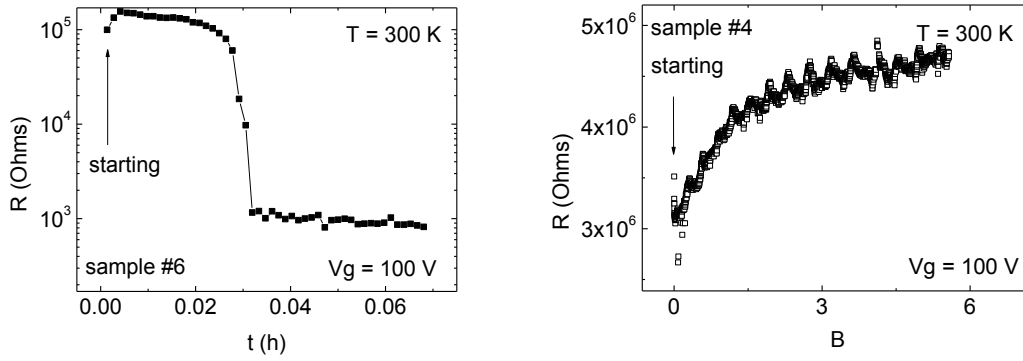


Fig. 2. Resistance vs. time monitored at fixed applied voltage $V_g = 100$ V for white (left panel) and partially oxidized greenish (right panel) CuCl samples.

It has been found that the transition to LRS is reversible. Figure 3 demonstrates the electric-field-driven transition between high- and low-resistance states measured in CuCl at $T = 300$ K, and its intermittent time dependence reducing V_g or changing the voltage polarity.

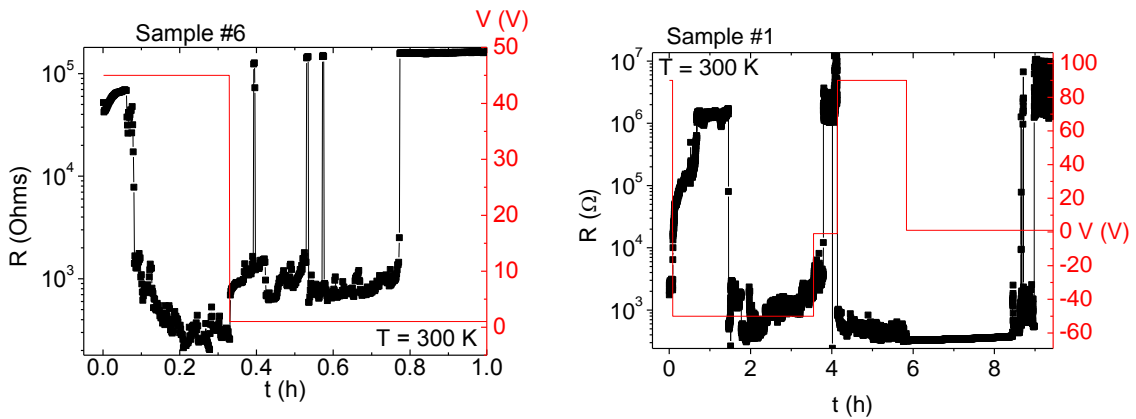


Fig. 3. Temporal switching between low- and high-resistance states measured at $T = 300$ K for two CuCl samples after reducing the applied voltage (left panel) or its sign reversal (right panel).

Measurements of the resistance vs. time at low enough temperatures revealed no transition towards LRS on the time scale of several hours. Figure 4 exemplifies this fact where measurements performed for the sample # 6 at $T = 3$ K revealed no transition during ~ 2 h whereas at $T = 300$ K, the transition takes place within several minutes (Fig. 3).

In samples made of a larger size particles, i. e. the samples that possess a smaller surface area, the resistance drop at the insulator-metal transition is very modest (factor ~ 4) implying that the conductivity in LRS is governed by the grain surface. Supporting this fact, we measured a similar voltage-driven HRS-LRS transition when two electrodes were placed on the same sample surface, i. e. in the “planar geometry”.

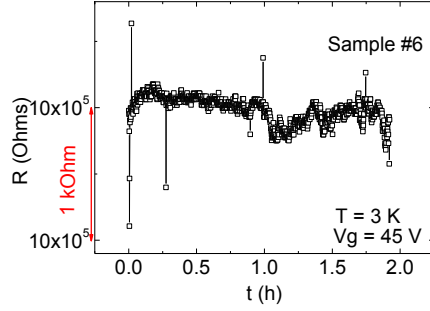


Fig. 4. Resistance vs. time monitored for the same sample (#6) at applied voltage $V_g = 45$ V and $T = 3$ K. Note the absence of the transition towards the low resistance state.

It is found that LRS is characterized by non-linear I-V characteristics, $I \sim V^\alpha$ with $\alpha = 3/2$ (Fig. 5), the behavior known for charge density wave (CDW) systems in a vicinity of the electric-field-driven (EFD) depinning transition [6]. The right inset in Fig. 5 provides evidence for the “metallic” ($dR/dT > 0$) origin of the conducting state. No metallic state was observed in four-probe measurements indicating that the metallic phase is localized within narrow quasi-one-dimensional (1D) channels.

One can speculate that applied at $T = 300$ K electric field $E > E_{th}$ (threshold field) delocalizes preexisting CDW leading to the highly conducting state. Alternatively, the low resistance state can be associated with the appearance of quasi-1D Cu filaments followed by a nucleation of CDW at Cu/CuCl interfaces.

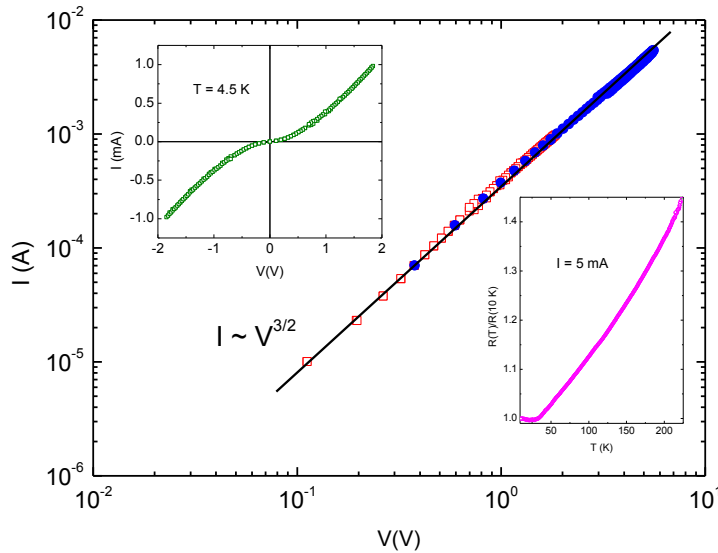


Fig. 5. Two-probe I-V characteristics measured for sample #6 at $T = 4.5$ K (\square) and $T = 4.15$ K (\circ). The solid line corresponds to the power law $I \sim V^{3/2}$. The left inset shows I-V curve measured at $T = 4.5$ K for both positive and negative applied voltage. The right inset presents a temperature variation of the reduced resistance $R(T)/R(10\text{K})$ in the electric-field-induced metallic state, measured at constant applied current $I = 5$ mA.

Assuming that sliding CDW form closed loops, strongly diamagnetic state accompanied by the resistance drop [1-3] can be understood without invoking the superconductivity in a usual sense. Instead, our results are consistent with the high-temperature Fröhlich superconductivity [7].

As temperature increases, I-V characteristics demonstrate more complex behavior related to the switching between metallic and insulating states (Figs. 6-7). The switching was observed after a certain number of I-V measuring cycles at a constant temperature. Certainly, a further experimental work is needed to shed light on the origin of this high-temperature metastable regime.

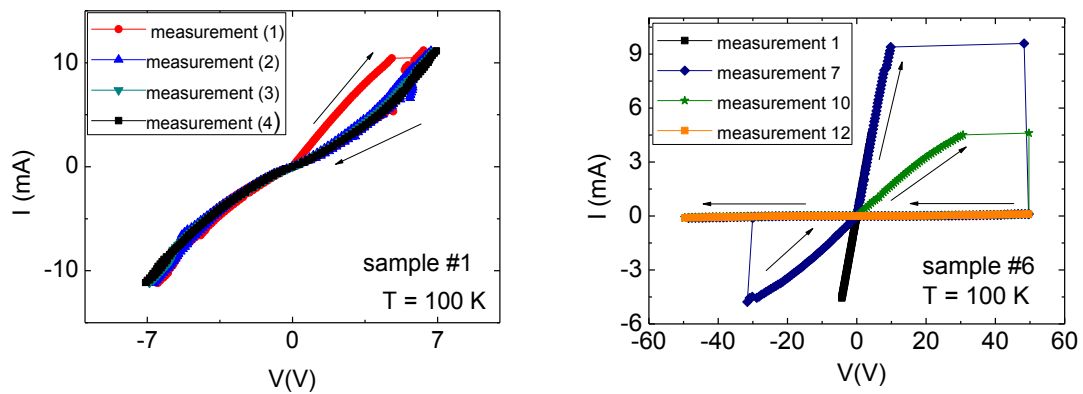


Fig. 6. Multiple I-V cycles measured for two samples at $T = 100$ K demonstrating hysteretic behavior and switching between low- and high-resistance states.

Low-temperature measurements in magnetic field applied either parallel or perpendicular to the measuring current and non-linear regime of I-V characteristics revealed a small ($\sim 0.1\%$ at $B \sim 7$ T) negative quadratic magnetoresistance, $\Delta R(B)/R \sim -B^2$, see Fig. 7. Similar magnetoresistance has been reported for α -TaS₃ CDW whiskers [8] and attributed to the delocalization of CDW solitons, supporting the Fröhlich-type CDW conductivity scenario.

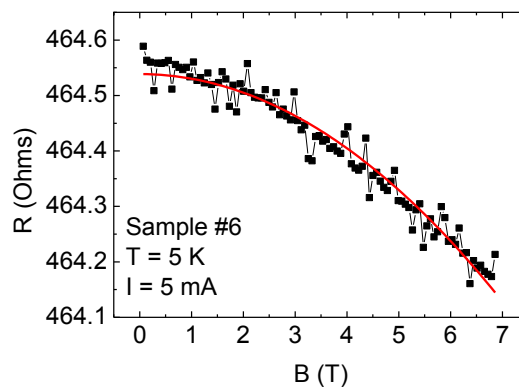


Fig. 7. Magnetoresistance measured for the sample #6 at $T = 5$ K and constant applied current $I = 5$ mA (non-Ohmic regime, see Fig.5). The red line corresponds to the parabolic fit to the data.

Besides of the electrical transport, we have performed magnetization $M(H,T)$ and electron spin resonance (ESR) measurements in both insulating and metallic states with no difference between the results. This comparative study corroborate our conclusion on the occurrence of a tiny (filamentary) metallic phase in the low resistance state of CuCl.

Summing up, the here reported results provide experimental evidence that the low resistance state in CuCl is governed by charge density wave motion along of some sort quasi-1D filaments. Electric-field-induced LRS, non-linear I-V characteristics, temporal fluctuations between low- and high-resistance states, aging effect, and negative parabolic magnetoresistance can be understood assuming the Fröhlich-type CDW (super)conductivity.

Part of the work described in this report is done in a collaboration with the University of Texas at Dallas (UTD). The obtained results are in preparation for publication.

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- [7] M. Tsubota et al., EPL 97 (2012) 57011 and refs. therein.
- [8] K. Inagaki et al., arXiv: 1407.6795.

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Dr. Iakov Kopelevitch

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Dr. Brett Pokines, AFOSR/IO, brett.pokines@afosr.af.mil

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Abstract

The results obtained within the framework of this project shed a new light on the long-standing problem of possible high- temperature superconductivity in CuCl. The obtained experimental evidence indicates that the low resistance state in CuCl is governed by motion of charge density wave(s) along quasi-one-dimensional filaments. The electric-field-induced metallic state, non-linear current-voltage characteristics, intermittent switching between low- and high-resistance states, aging effect, and negative parabolic magneto-resistance, all these can be understood assuming the Fröhlich-type (super)conductivity in the studied material.

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