Supporting Information file

Cationic order-related magnetoresistivity and half-metallicity in bulk Pb_2FeMoO_6 grown by high pressure synthesis

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S1. Transport Models

In order to investigate the nature of PFMO resistivity, we have considered different transport models for both the datasets measured with H=0 T and H=2 T and reported in Figure 1 of the main text. Namely, we have evaluated the resistivity trend as a function of temperature with the Thermal Activated (TA) transport, the Adiabatic nearest Neighbors Hopping of Small Polarons (ANHSP), and the Mott variable-range hopping mechanism (VRH) whose equations are reported in eq. 1-2-3 respectively:

$$\rho = \rho_0 \cdot exp\left(\frac{E_A}{kT}\right) \quad (1)$$

$$\rho = \rho_0 T \cdot exp\left(\frac{E_{HOP}}{kT}\right) \quad (2)$$

$$\rho \approx \rho_0 \cdot exp\left(\frac{T_0}{T}\right)^{1/(D+1)} \quad (3)$$

with k as the Boltzmann constant. For TA (1), E_A is the activation energy and ρ_0 is the value of resistivity at infinite temperature. For ANHSP (2), the effective resistivity is defined as $\rho_0 = 2k/(3ne^2a^2v)$, where n is the density of charge carriers, e is the electronic charge, a is the hopping spatial distance and v is the longitudinal optical phonon frequency. E_{HOP} is the hopping threshold energy of the transport process. In the Mott variable-range hopping (3), the D parameter specifies the dimensional conductance and can be equal to 1, 2, or 3 along one, two or three directions.

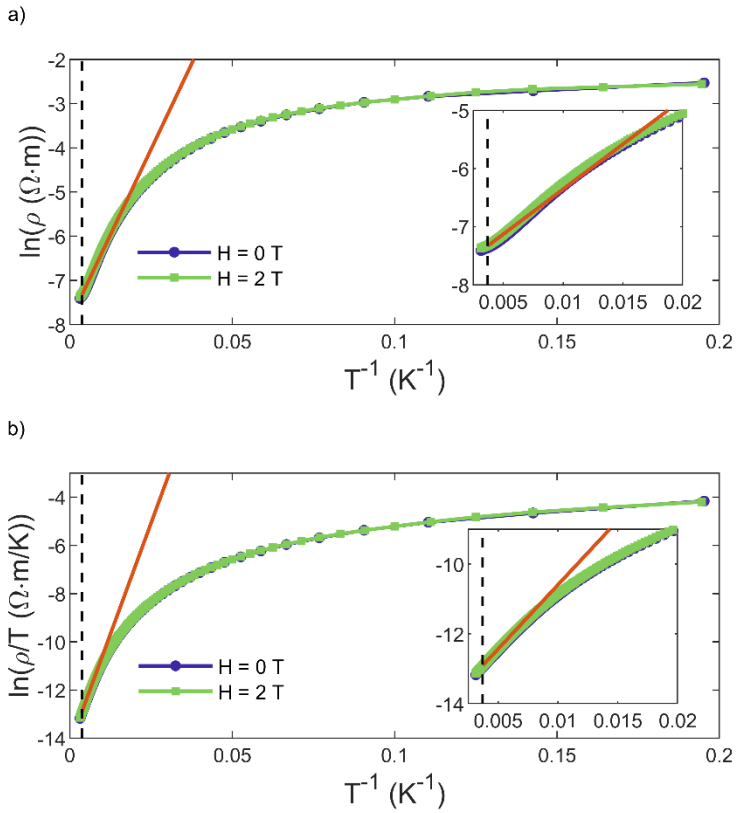

Figure S1. Resistivity as a function of temperature considering (a) the Thermal Activated transport or (b) the Adiabatic nearest Neighbors Hopping of Small Polarons transport models. The zoom of the data is reported in the inset. The dashed black line corresponds to the T_N.

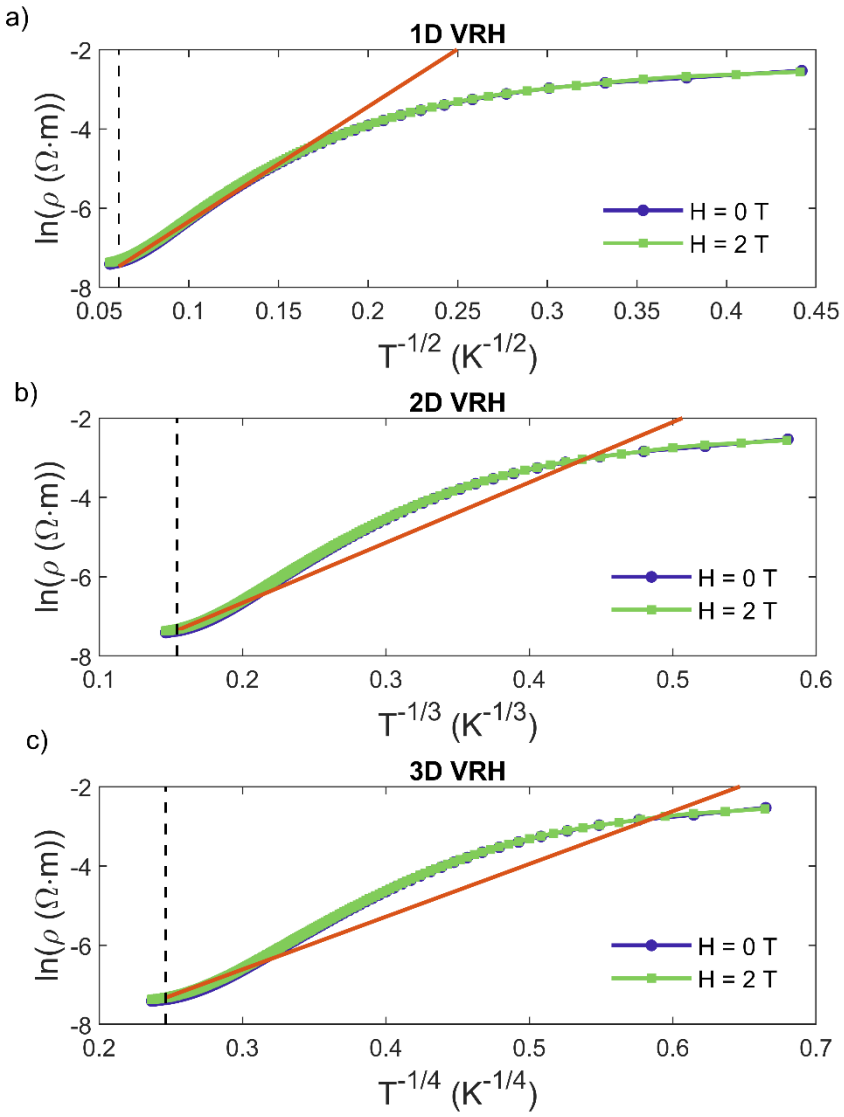

Figure S2. Resistivity as a function of temperature considering (a) 1D, (b) 2D or (c) 3D Variable range Hopping mechanism. The zoom of the data is reported in the inset.

In Figure S1-S2 all the models have been reported and is clearly visible that neither model describes the resistivity behavior vs. temperature in the low T ferrimagnetic state, where non-zero values for the first derivative of ρ have been calculated and a non-exponential trend is recorded, with a saturation of ln(ρ) at low temperature. As a consequence, we can state that our PFMO does not display semiconductor behavior in the ordered magnetic state below T_N.

As none of the previous models has been effective for the modelling of PFMO resistivity, the additional fluctuation-induced tunnelling (FIT) model, reported to well explain the electrical transport of the parent compound Sr_2FeMoO_6 and, particularly, polycrystalline ceramics [1], has been considered.

FIT is a three parameters-thermally activated law with a shift in temperature of the exponential divergence from 0 K towards an arbitrary temperature value T_0:

$$\rho = \rho_0 \, e^{\frac{T_1}{T+T_0}}$$

In Figure S3 the fitting of the resistance dataset recorded without the magnetic field applied with this model is shown. The results, considering the whole thermal range, apparently display a good match between experimental data and the FIT function with high $R^2 = 0.9992$. The fit returns $T_1 = 599$ K, $T_0 = 79$ K and $R_0^* = 6.25 \cdot 10^{-5}$ Ω. However, if a zoom in the flat region of the curve is performed, for example in the thermal range between 120 K and 325 K, as reported in the inset of figure S3, a systematic deviation of the fitting curve from the experimental data is clearly visible, which becomes more and more affected by the error as the temperature is increased, demonstrating that the model does not fit the whole curve.

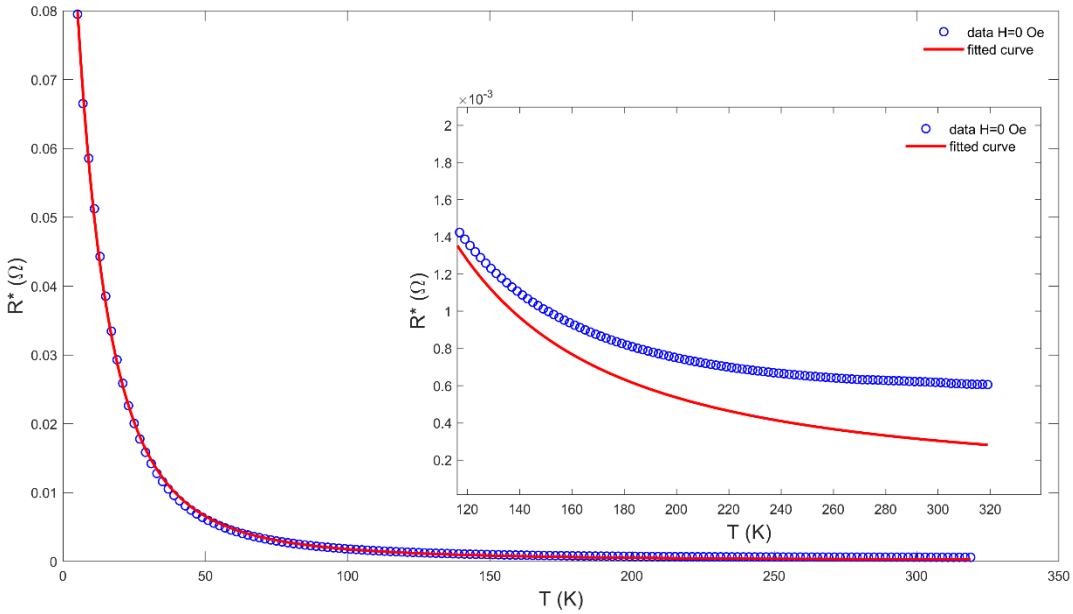

Figure S3. Resistance vs T trend evaluated with FIT model. In the inset, the zoom on the thermal range between 120 K and 320 K underlines the discrepancy between the data and the considered model.

S2. Magnetoresistance

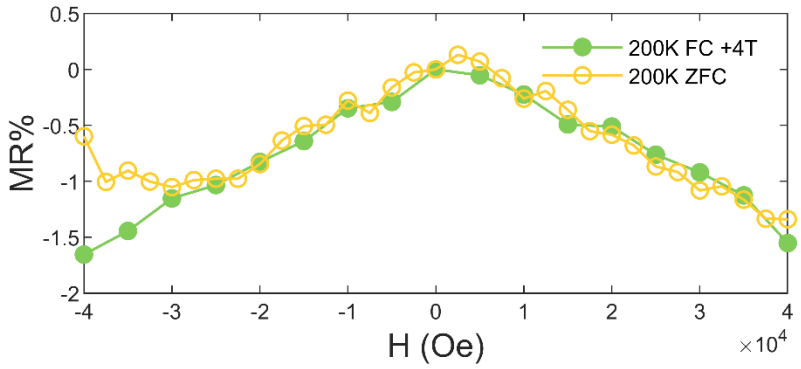

Figure S4. Magnetoresistance measurements collected between –40 kOe and 40 kOe at 200 K after ZFC (empty dots) and applying +40 kOe after FC (filled dots).

In the case of the measurement performed after 40 kOe FC protocol (Figure S4, filled dots) a standard symmetric magnetoresistive effect is measured with a maximum relative loss of about 1.5% at –40 kOe and 40 kOe. In this case, the effect of a strong positive magnetic field during the cooling acts in a reduction of the conduction, as observed in the transport characterization, thus avoiding the promotion of the polarized carriers. A completely different phenomenology has been observed in the case of a negative magnetic field applied during the cooling process (Figure 3a), as described in the main text file.

References

[1] Suchaneck, G. and Artiukh, E., Absence of Weak Localization Effects in Strontium Ferromolybdate, Applied Sciences, 13, 12, 2023 (doi: 10.3390/app13127096).