Supplementary Materials: Anomalous Pressure Effects on the Electrical Conductivity of the Spin Crossover Complex [Fe(pyrazine){Au(CN)$_2$}$_2$]

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Experimental details

Magnetic measurements were performed using a MPMS3 SQUID magnetometer (Quantum Design Inc.), in DC mode, under a DC magnetic field of 1000 Oe, with a temperature rate of 2 K/min. Data were corrected for the sample holder and sample diamagnetic contributions. High-pressure magnetic data were acquired using a clamp-type cell calibrated by the superconducting transition of Pb.

Electrical impedance measurements have been performed with a CONCEPT 40 Broadband Dielectric Spectrometer (Novocontrol GmbH) as a function of temperature under constant pressure. The spectra have been recording with an Alpha-A high performance frequency analyzer (3 μHz…20 MHz) and a High Pressure System option for dielectric measurements from 0 to 3 kbar in the 25 °C – 250 °C temperature range. The temperature was changed in sweeping mode at 0.5 K/min, in both heating and cooling modes, and the pressure was manually lowered (increased) to maintain a constant value (±5 bar) throughout the entire temperature sweep range, while continuously recording frequency spectra.

IR spectra were recorded with a Perkin Elmer Spectrum Two FTIR spectrometer in attenuated total reflectance (ATR) mode in ambient conditions.

Scanning Electron Microscopy (SEM) micrographs were recorded using a Hitachi SU-70 microscope. The particles have been deposited on an Al mount from a previously sonicated suspension in toluene.

Elemental analysis:
S1 C8H4Au2FeN6 (633.9): calcd. C 15.16, N 13.26, H 0.64; found C 15.44, N 13.22, H 0.44.
S2 C8H4Au2FeN6 (633.9): calcd. C 15.16, N 13.26, H 0.64; found C 15.55, N 13.54, H 0.48.

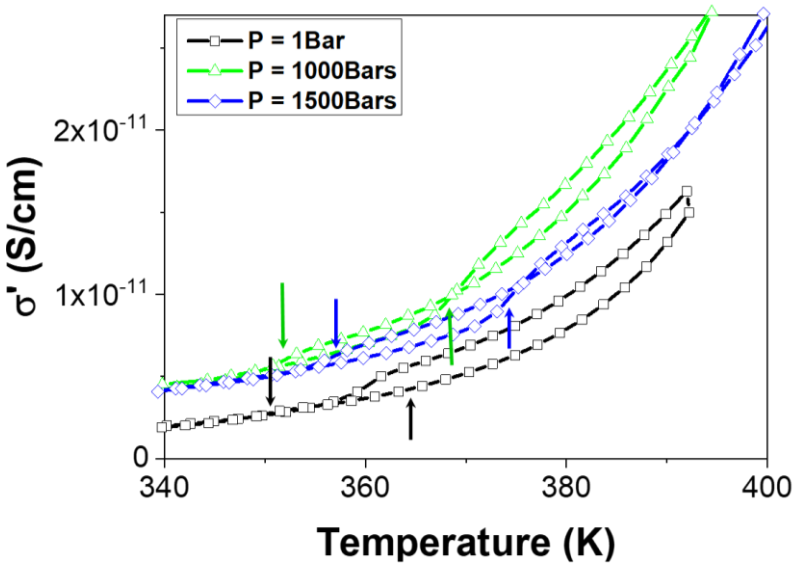

Figure S1. Temperature dependence of the ac electrical conductivity recorded on **S2** under various external pressures. The arrows indicate the corresponding switching temperature.

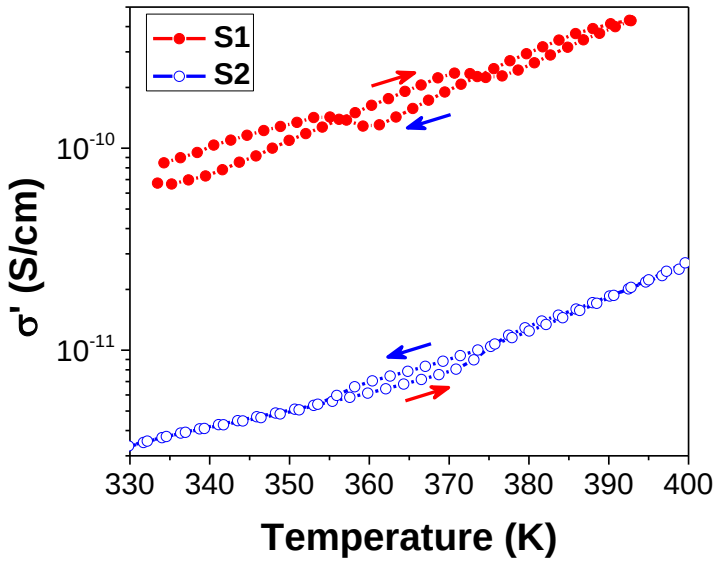

Figure S2. Temperature dependence of the ac electrical conductivity (f=100 Hz) recorded on S1 and S2 under an external pressure of 1500 bar.

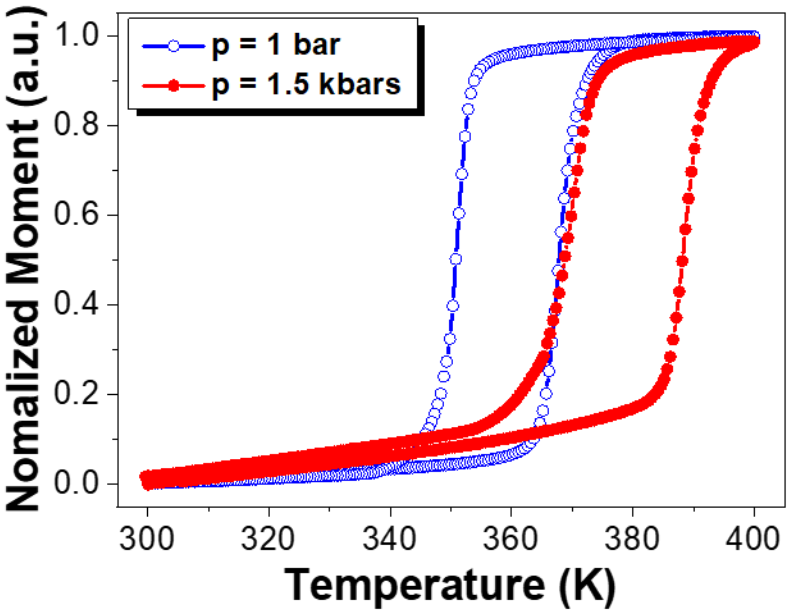

Figure S3. Temperature dependence of the molar high spin fraction recorded from magnetic measurements on S1 under two various external pressures.

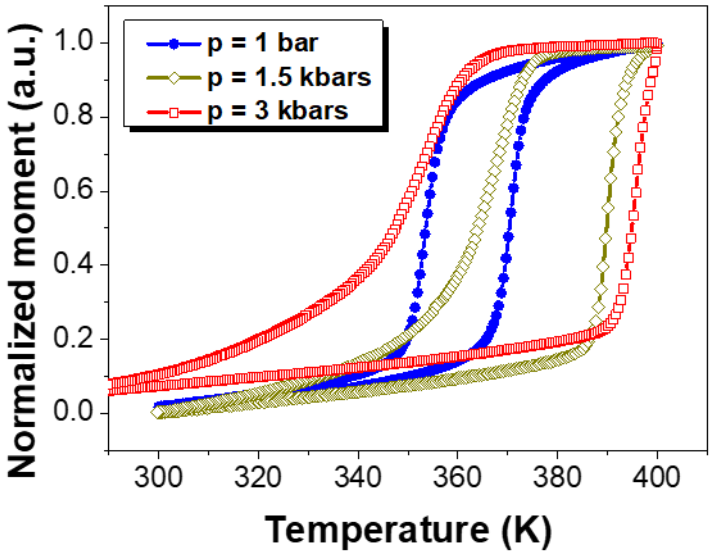

Figure S4. Temperature dependence of the molar high spin fraction recorded from magnetic measurements on **S2** under various pressures.

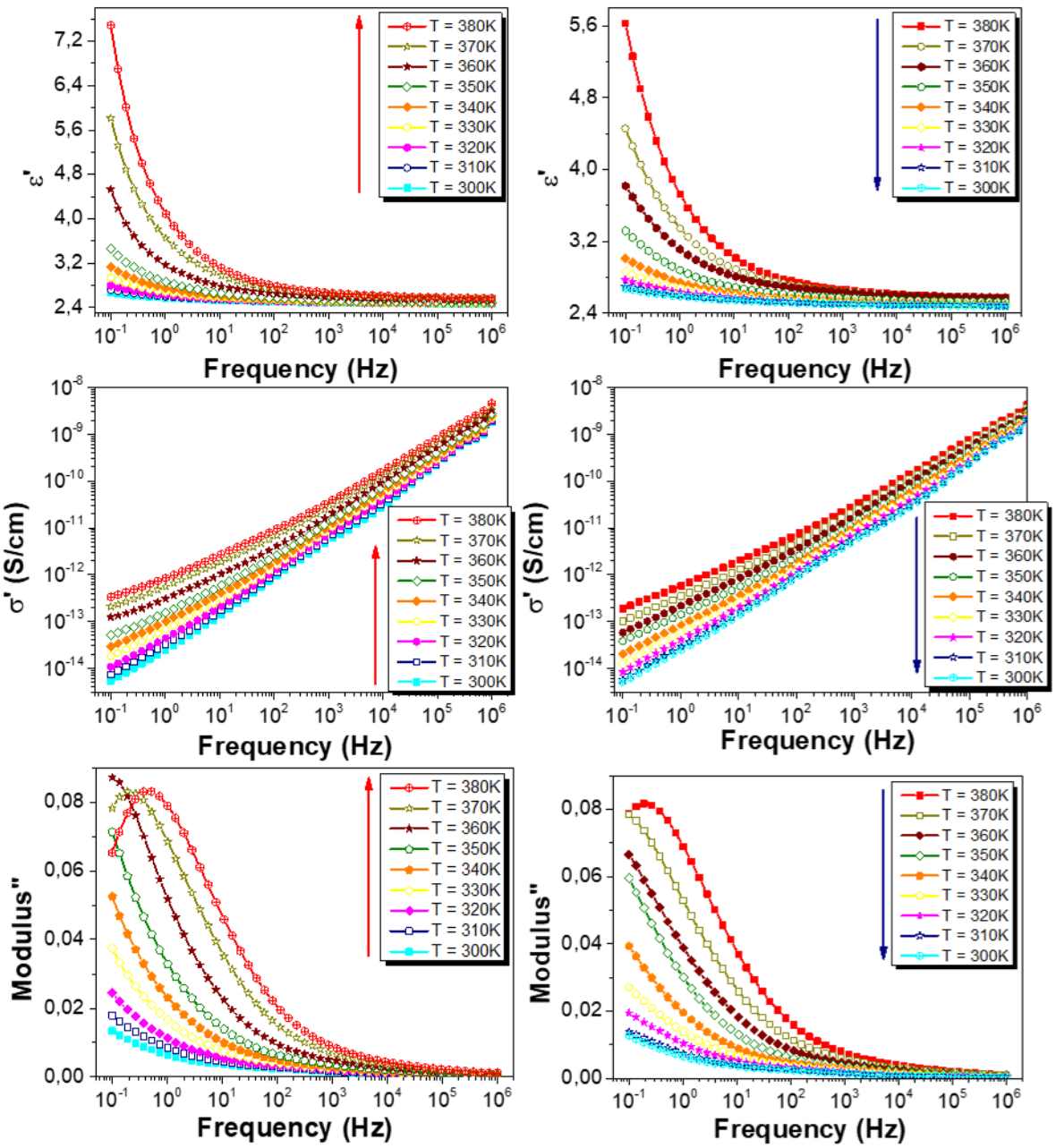

Figure S5. Frequency dependence of the real part of the dielectric permittivity, real part of the ac conductivity and imaginary part of the electrical modulus recorded on **S1** at ambient pressure at various temperatures values in heating (Left) and cooling (Right) modes.

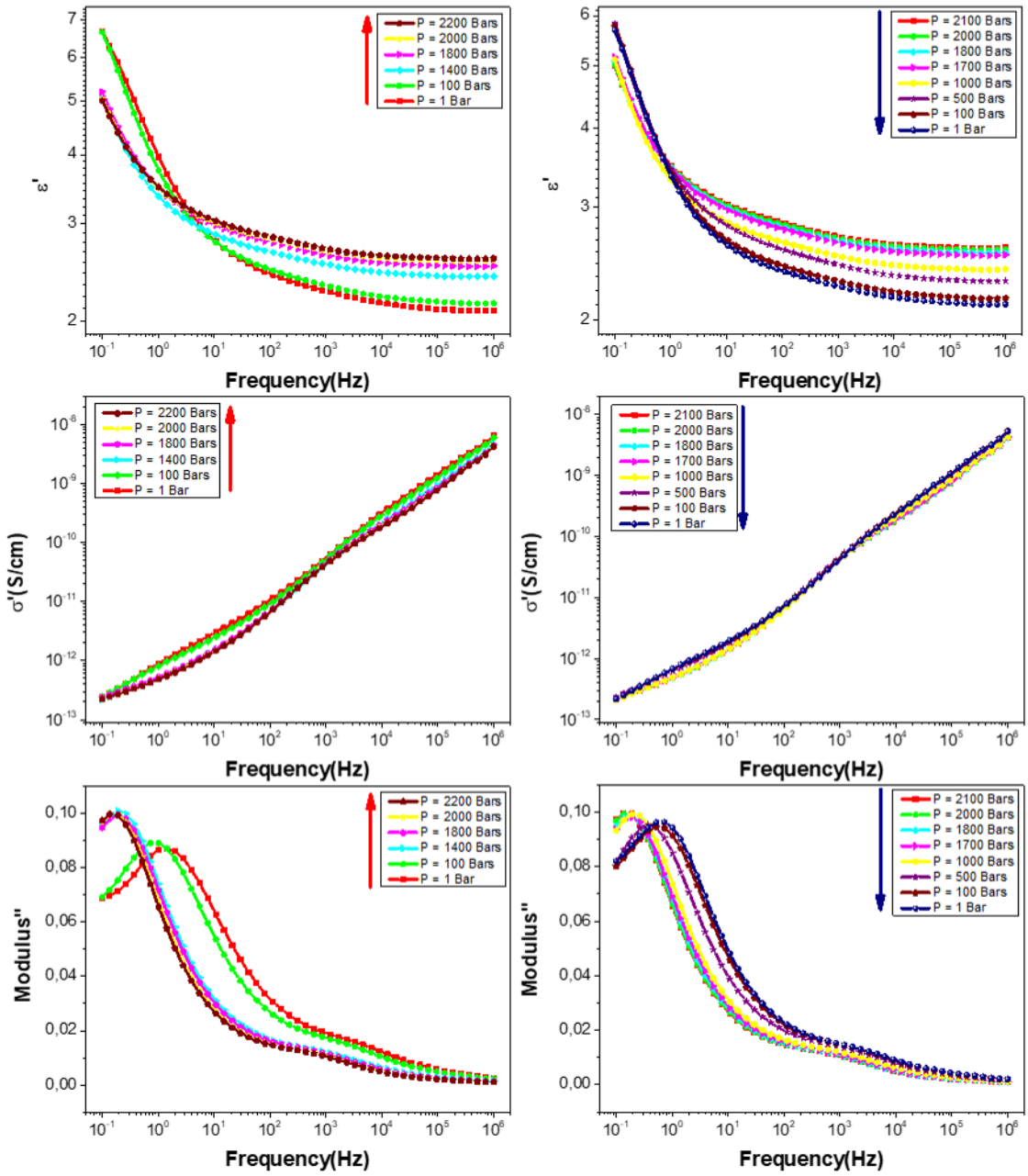

Figure S6. Frequency dependence of the real part of the dielectric permittivity, real part of the ac conductivity and imaginary part of the electrical modulus recorded on **S1** at 368 K for various pressure values in the loading (Left) and unloading (Right) modes.

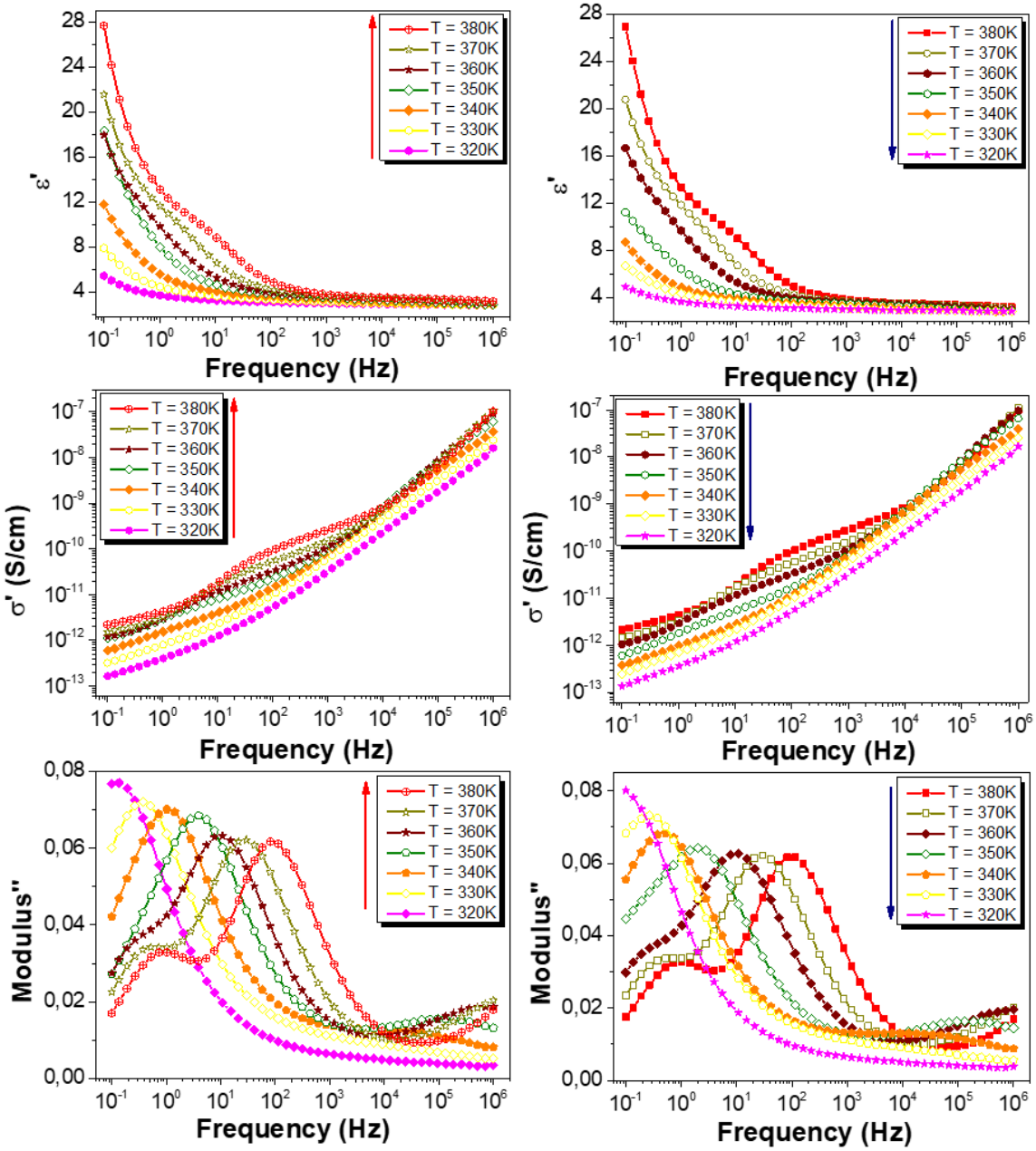

Figure S7. Frequency dependence of the real part of the dielectric permittivity, real part of the ac conductivity and imaginary part of the electrical modulus recorded on **S2** at ambient pressure at various temperatures values in heating (Left) and cooling (Right) modes.

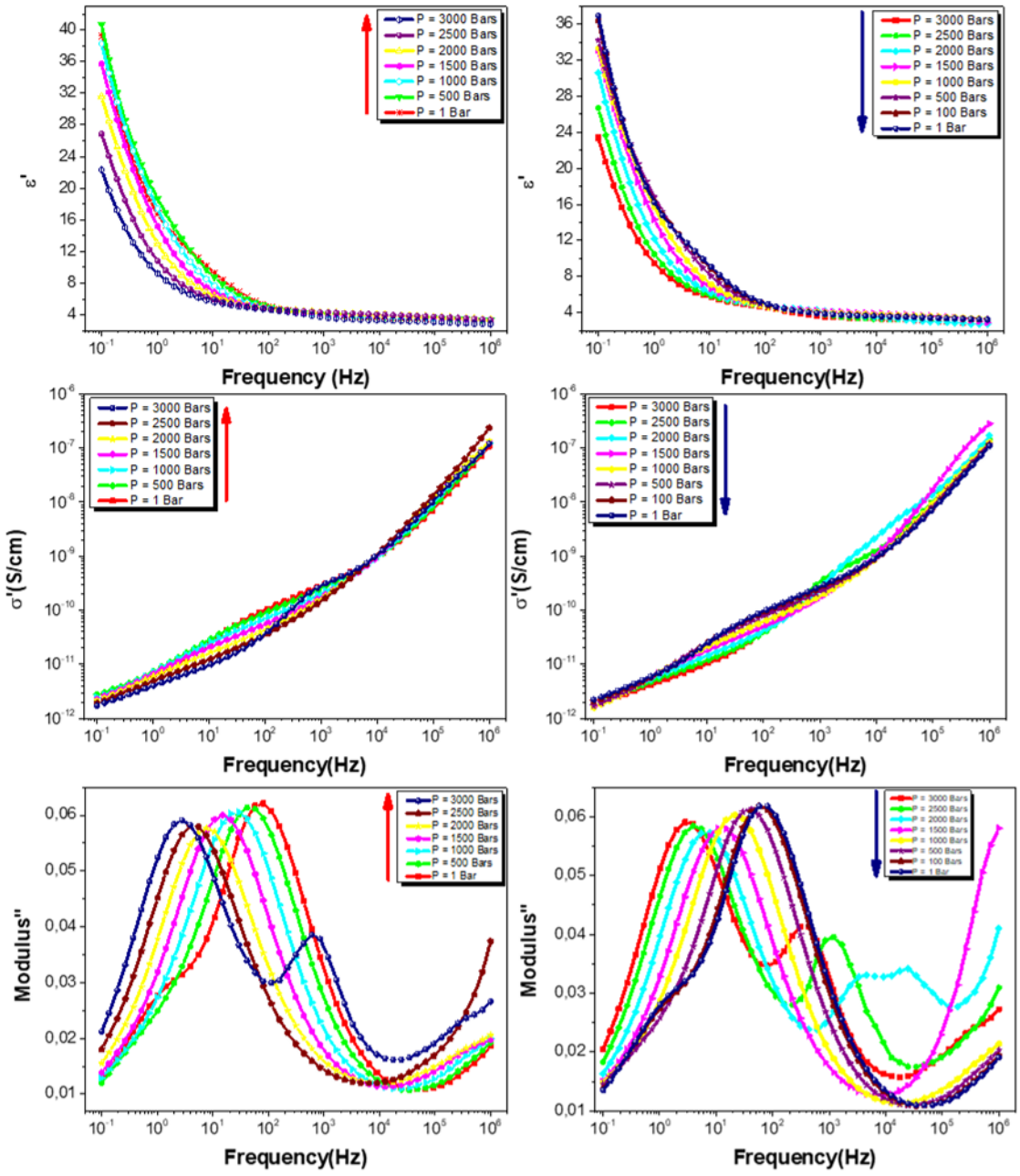

Figure S8 Frequency dependence of the real part of the dielectric permittivity, real part of the ac conductivity and imaginary part of the electrical modulus recorded on **S2** at 373 K for various pressure values in the loading (Left) and unloading (Right) modes.

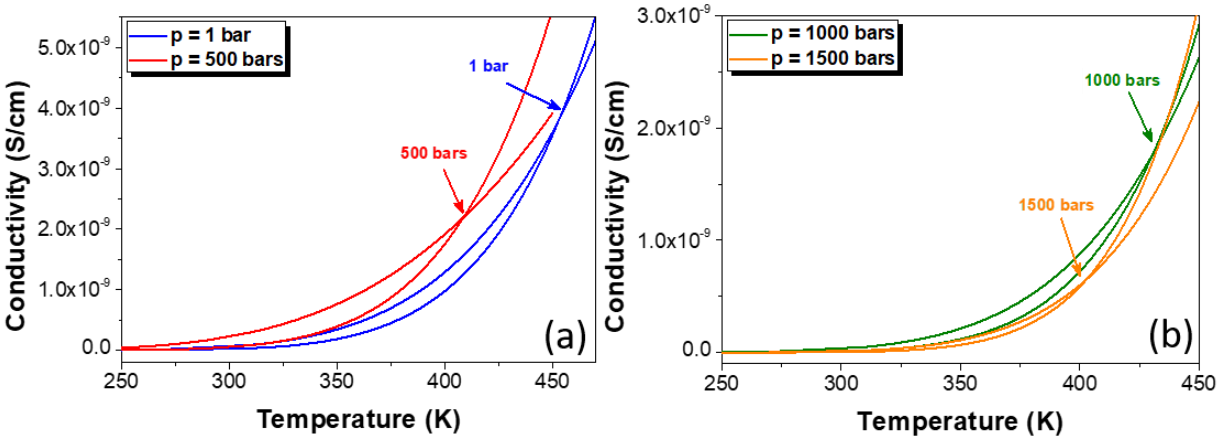

Figure S9. Temperature dependence of the electrical conductivity simulated by using the fitting parameters resulted from the analysis of **S2** from Table 1, at various pressures: **(a)** 1 bars and 500 bars and **(b)** 1000 bars and 1500 bars.