Dielectric anisotropy of the organic crystals $\theta$-(BEDT-TTF)$_2$M[Zn(SCN)$_4$]$_2$ and $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl

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The organic layered crystals $\theta$-(BEDT-TTF)$_2$M[Zn(SCN)$_4$]$_2$ (M=Cs,Rb) and $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl show highly nonlinear current-voltage (I-V) characteristics in a low current region [1,2]. The nonlinear I-V curves are explained in terms of electric-field-assisted unbinding of electron-hole pairs, which are thermally excited in the charge-order or in the Mott insulating state. The analysis of the I-V curves indicates a two-dimensional (2D) long-range Coulomb interaction, logarithmic in the distance between the electron and the hole.

In this study, we measured low-frequency (1 Hz – 100 kHz) dielectric constants along both the in-plane and out-of-plane directions at low temperatures (≥ 0.4 K). The anisotropy in the dielectric constant of M=Cs is as large as ≈100 in the low-temperature limit. This value corresponds to the cut-off length of ≈20 nm of the 2D Coulomb interaction, which is on the same order of the value estimated from the nonlinear I-V curves. The temperature and frequency dependencies of the in-plane dielectric constant are explained by the polarization of the bound electron-hole pairs, consistently with the nonlinear I-V curves. We also report on dielectric anisotropy of $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Cl.