Infrared and Raman studies of quasi-one-dimensional conductors $(o\text{-DMTTF})_2X$ (X = Br, I)

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The quasi-one-dimensional organic metals formed by the organic donor 3,4-dimethyl-tetrathiafulvalene (*o*-DMTTF) with Br^- or I^- anions possess strictly uniform *o*-DMTTF stacks down to the lowest temperatures. Organic conductors with uniform stacks are rare and allow investigation of electron-electron correlations in one dimension, i.e. the problem of Mott localization in a one-dimensional 1/4 filled band (in terms of holes). The studied compounds are isostructural and their characteristic feature is the presence of hydrogen bonds between methyl groups and halide anions. The salt $(o\text{-DMTTF})_2Br$ undergoes a metal-insulator phase transition at T_{MI} = 50 K [1].

Polarized IR reflectance spectra (700 – 14000 cm^{-1}) and Raman spectra (excitation λ = 632.8 nm) of $(o\text{-DMTTF})_2X$ (X = Br, I) single crystals were measured as a function of temperature (T = 4 – 300 K). Using Kramers-Kronig relation the optical conductivity spectra were calculated from the reflectance data. Additionally, IR and Raman spectra of neutral *o*-DMTTF were measured at room temperature. The normal vibrational modes of *o*-DMTTF molecule were calculated by DFT method.

The conductivity spectra of both salts are very similar and exhibit an optical gap around 5000 cm^{-1} at room temperature, despite the metallic behavior found from the DC conductivity. The metal-insulator transition in Br salt at 50 K has nearly no influence on electronic dispersion but some minor modifications are seen at about 200 K. IR vibrational features recorded for polarization perpendicular to the stacks were also analyzed for different temperatures. In Raman spectra three strong lines related to the C=C stretching modes were observed at 1480, 1509, 1600 cm^{-1} for Br salt and 1475, 1510, 1601 cm^{-1} for I salt. Temperature dependence of these Raman bands shows that in both salts the charge distribution in *o*-DMTTF stacks is uniform in the whole temperature range; there is no charge ordering in Br salt below T_{MI} = 50 K. The spectra of Br and I salts were compared and some differences discussed. Additionally, the theoretical and experimental vibrational spectra obtained for *o*-DMTTF molecule (IR and Raman) were analyzed and discussed in relation with analogous spectral data for TTF and TMTTF molecules.

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[1] M. Fourmigué *et al.*, Dalton Trans. 2008, 4652.