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α-(BEDT-TTF)2I3: Complex electrodynamic response of the charge-ordered phase

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Much experimental and theoretical attention has been attracted by organic systems with reduced dimensionality and strong Coulomb interactions, and deservedly so due to their novel broken-symmetry phases and corresponding excitations. Here we take a detailed look at the electrodynamics of one of the most prominent charge-ordered systems, the quasi-2D conductor α-(BEDT-TTF)2I3. A semimetal at high temperatures, at 136 K this particular system transitions into an insulating, diamagnetic ground state. Within the insulating phase a long-range commensurate ordering appears in the BEDT-TTF molecular planes, the so-called "horizontal stripe" charge order. [1,2,3] We characterize the charge response of the low-temperature phase using dc resistivity, dielectric and optical spectroscopy in different crystallographic directions within the BEDT-TTF layer. [4,5] Interestingly, two dielectric relaxation modes appear in the kHz-MHz range. The large mode features an anisotropic phason-like behavior, while the small mode presents a soliton-like characteristic. The observed type of excitations agrees with the most relevant physical picture of this charge order as a cooperative bond-charge density wave with ferroelectric-like features. [6] On the other hand, puzzling phenomena including negative differential resistance and voltage oscillations have been reported under application of high electric fields. [7] Our carefully designed electric-field-dependent measurements of conductivity anisotropy within the molecular plane qualitatively confirm these findings; additionally, they reveal novel intriguing behaviors. [8].

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