Frustration and Lattice Effects on Photoinduced Melting of Charge Orders in Quasi-Two-Dimensional Organic Conductors

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To elucidate different photoinduced melting dynamics of quite similar, horizontal-stripe charge orders observed in quasi-two-dimensional organic conductors θ-$(BEDT\text{-}TTF)_2RbZn(SCN)_4$ (abbreviated as θ-RbZn) and α-$(BEDT\text{-}TTF)_2I_3$ (as α-I_3) [1], we theoretically study photoinduced evolution of the wave functions in extended Peierls-Hubbard models on anisotropic triangular lattices. The exact many-electron wave function coupled with classical phonons or the unrestricted Hartree-Fock approximation is used depending on the system size. These charge orders are stabilized by both Coulomb repulsion and electron-lattice interactions. Their relative importance is different between θ-RbZn and α-I_3 [2,3].

In θ-RbZn, a large lattice distortion is necessary to lift the degeneracy among many charge-ordering patterns, which are possible owing to frustration caused by the high structural symmetry. The lattice stabilization energy is consequently large, and a high photoexcitation density is required for the melting. Each hole-rich stripe is stabilized by molecular rotation as a whole, so that the "intra-stripe" ("inter-stripe") charge correlation is strong (weak). This results in anisotropic growth of a photoinduced metallic domain after local photoexcitation. The photoinduced charge dynamics shows a complex behavior owing to a large number of nearly degenerate eigenstates involved owing to the frustration.

In α-I_3, a small lattice distortion is sufficient to stabilize the charge order because the low structural symmetry ensures partial charge disproportionation in the metallic phase. The lattice stabilization energy is small, and a low photoexcitation density is sufficient to melt the charge order producing a metallic phase. Because each hole-rich bond is locally stabilized, even the "intra-stripe" charge correlation is weak. This results in more isotropic growth of a photoinduced metallic domain after local photoexcitation. The photoinduced charge dynamics shows a coherent oscillation when resonantly excited.

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