Inelastic neutron scattering studies on low-energy molecular lattice dynamics in organic conductors

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We report the low energy phonon dynamics coupled with charge and spin degrees of freedom in molecular charge transfer salts studied by measuring inelastic neutron scatterings (INS). A π-electron system in molecular conductors inherently involves characteristic charge-spin-lattice degrees of freedom in the hierarchical structure of electrons in the molecule (spins and charges), intra-molecule coupling (molecular vibrations) and inter-molecule coupling (molecular lattice phonon). The multi-degrees of freedom combined with the structural flexibility of molecular materials induce novel dynamical spin and charge states. Recent findings of spin and charge degrees of freedom (spin liquid and charge glass states) in the π-electron system should have close connection to the lattice dynamics through the coupling between the π electrons with S=1/2 spin and σ electrons which form the intra-molecular bonding. INS is powerful probe for observing low energy phonon mode. But there have been few reports on INS experiment in molecular organic materials [1] because of less available volume of the crystals and containing large amount of hydrogens.

Recently, A molecular dimer-Mott insulator β'-(BEDT-TTF)$_2$ICl$_2$ is found to show a new type of electric field induced ferroelectric state coupled with the antiferromagnetic spin correlations and the long range order [2]. We have performed INS measurements on this system by using a cold-neutron disk-chopper spectrometer AMATERAS, J-PARC, and observed phonon spectra. Clear optical modes were observed together with acoustic phonon dispersion. The intensity of the optical mode with the lowest energy 4.2 meV increases at a zone boundary below 62 K at which charge disproportionation is expected to occur. The intensity of this mode increases especially for the zone boundary modes at (0k0) with half integer k below T_N = 22 K. The results suggest that the lattice dynamics characterized by this optical phonon mode is coupled with strongly correlated charge and spin degrees of freedom.

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