Synchronized Charge Oscillations in Conductors with Dimer Structures

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In an organic superconductor κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br, which is located near the Mott criticality, stimulated emission [1] and second harmonic generation (SHG) [2] are experimentally observed. The former is theoretically shown to be caused by the emergence of an electronic breathing mode (Fig. 1) as a nonlinear charge oscillation that appears only after strong photoexcitation [3]. Here, I discuss that synchronization of charge oscillations is necessary to observe such stimulated emission and that the responsible oscillation is also related to SHG in this centrosymmetric material. These results are obtained by using the exact diagonalization method and numerical solutions to the corresponding time-dependent Schrödinger equation.

After the application of a very short (~1 cycle) pulse of a specifically polarized electric field, the charge densities at half of the molecules oscillate in the same phase and those at the other half oscillate in the opposite phase. For strong fields, the peaks in the optical conductivity spectrum are suppressed and a new peak appears on their high-energy side in the Fourier spectrum for the charge density at a molecule. That is, the charge densities mainly oscillate with a single frequency, although the oscillation is eventually damped. This frequency corresponds to charge-transfer processes by which all the bonds connecting the two classes of molecules (characterized by different transfer integrals) are exploited. Thus, this oscillation behaves as an electronic breathing mode [3]. It is found that the on-site Coulomb repulsion is needed to synchronize the charge oscillations and it overcomes numerically introduced randomness in transfer integrals (Fig. 2) to artificially suppress the synchronization [4]. The synchronization mechanism is understood on the basis of the Bloch equation for pseudospins [5]. Furthermore, such a strong optical field can bring the system into a negative-temperature state [3,4], which is consistent with the experimental observation of stimulated emission (i.e., the many-body version of population inversion).

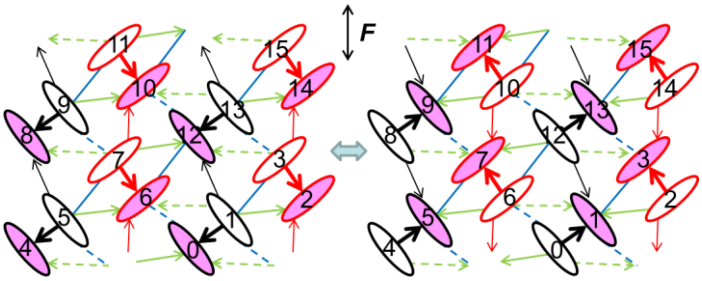

Fig. 1. Electronic breathing mode.

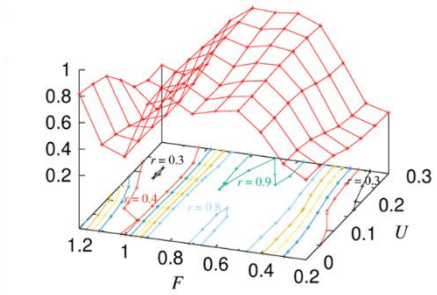

Fig.2 Synchronization order parameter as a function of on-site repulsion U and field strength F for randomness $\varepsilon = 0.3$.

References

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