Ultrafast spin/charge dynamics in quantum-spin materials: Symmetry aspects

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A change in the symmetry of an electron system by application of a short and strong optical field is a main issue in photoinduced phase transitions and cooperative phenomena. In the past, lowering the symmetry was often regarded as spontaneous or described with a probability. However, the progress in experimental techniques allows designed lowering of the symmetry, which is described deterministically, by applying a force to many electrons instantaneously before relaxation becomes significant. In this presentation, I theoretically discuss the mechanisms of pseudospin polarization in the quantum liquid state of α-$RuCl_3$ (time reversal symmetry breaking) and second harmonic generation in centrosymmetric κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br (space inversion symmetry breaking), both of which are achieved by S. Iwai's experimental group.

Pseudospin polarization in α-$RuCl_3$: This compound is known to show a quantum spin liquid state above T_N, which is basically described by the Kitaev model. T. Amano *et al.* observed the inverse Faraday effect during the application of circularly polarized light [1]. For systems with spin-orbit couplings, circularly polarized light is known to polarize spins (for instance, [2]). Here, we use an itinerant electron model and parameters employed in [3] (but the hopping is limited to the nearest neighbors), where three t_{2g} orbitals on a honeycomb lattice (Fig. 1) have strong on-site interactions of Kanamori type and strong spin-orbit coupling. The exact diagonalization method is applied to a system of six sites with periodic boundary conditions maintaining threefold symmetry. Numerical solutions to the time-dependent Schrödinger equation are analyzed with the help of a high-frequency expansion of quantum Floquet theory.

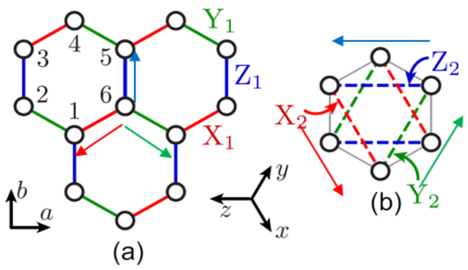

Figure 1: Honeycomb lattice showing **(a)** first and **(b)** second neighbors.

The high-frequency expansion shows two types of effective magnetic fields during the application of a circularly polarized light field. One of them is of purely kinetic origin and perpendicular to the lattice, thus producing nonzero and helicity-dependent perpendicular components of the pseudospins and consequently that of magnetization. The other originates from strong spin-orbit coupling and rotates almost within the honeycomb lattice according to the momentum distribution of holes. Both dynamics are confirmed numerically when spin-orbit-coupled in-gap states are excited below the Mott gap. The former is consistent with the finding in the experiment [1].

Second harmonic generation (SHG) in κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br: This *centrosymmetric* compound is known to show superconductivity and located near the Mott critical point. Y. Kawakami *et al.* observed SHG, which is sensitive to the carrier envelope phase [4]. If time allows, I show the mechanism based on [5] and [6]. They are understood from the viewpoint of current-induced SHG.

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